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ARCTIC HAZE: NATURAL OR POLLUTION?

A Progress Report and Renewal Proposal to the

Office of Naval Research

Arctic Program

1 September 1977

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Graduate School of Oceanography
University of Rhode Island
Kingston, Rhode Island 02881

and

Geophysical Institute
University of Alaska
Fairbanks, Alaska 99701

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DATE: 1 September 1977

AGENCY: Office of Naval Research - Arctic Program

Submitted

by

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UNIVERSITY OF RHODE ISLAND

KINGSTON, RHODE ISLAND

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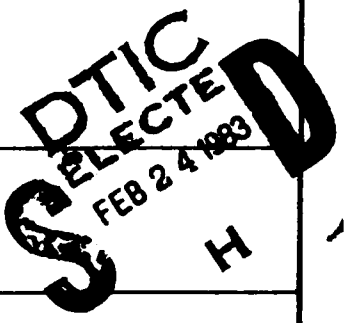

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20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*

The research reported here has: (1) Confirmed by comprehensive neutron activation analysis that the haze aerosol of April and May 1976 was crustal rather than pollution in origin. These bands could not have come from Europe or from the northeastern United States. (2) Further confirmed the crustal-dust nautre of these bands by scanning electron microscopy and electron-microprobe chemical analysis. (3) Confirmed by trajectory analysis that air from eastern Asia reached Barrow during the times that haze bands were there in April and May 1976. (4) Identified a major dust storm in

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eastern Asia at the proper time and place to have been ths source of the haze bands. This storm was followed on surface weather maps for nearly 3000 km over the Asian mainland, Korea, and Japan, until it was out over the Pacific. (5) Identified pollution aerosol in the surface atmosphere of Barrow, Alaska during winter. Much of this pollution is probably derived from mid-latitudes via long-range transport, and is seen at Spitsbergen as well. (6) Posed a meteorological hypothesis to explain the large winter-summer differences in pollution-aerosol concentrations in Barrow. (7) Noted the similarity between winter pollution maxima and spring maxima of Arctic turbidity, and tentatively identified the unusually high Arctic turbidities with pollution aerosol. In spring (and probably also winter), Arctic turbidities appear to be significantly higher than subarctic turbidities.

ARCTIC HAZE: NATURAL OR POLLUTION?

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Statement of Submission

The following progress report and renewal proposal, "Arctic Haze: Natural or Pollution?", is hereby submitted to the Office of Naval Research, Arctic Program, for consideration as a research contract. The proposal is complete except for the approval sheet from the University of Alaska, which should arrive within a week of the basic proposal.

This proposal is not being submitted to any other agency for financial support, although certain costs are to be shared with a ~~new~~ existing grant from proposal to the National Science Foundation ("Climatically Important Properties of Arctic Haze", Division of Polar Programs).

ABSTRACT

Our studies of Arctic haze aerosol to date have revealed that haze layers over Alaska can be composed of Asian desert dust; they have also suggested that a more pervasive diffuse haze may be pollution-derived. In order to expand our meager data base we will continue both ground-level and airborne studies this year.

Ground-level aerosol sampling will be continued at Barrow and Fairbanks; samplers may be added in Korea, New York City, and Rhode Island to better determine seasonal source functions for Asian dust and pollution aerosol, respectively. Samples from Greenland, northern Norway, Bear Island, and Spitzbergen will also be provided to this project by Scandinavian scientists. All samples will be analyzed for trace elements by neutron activation.

An intensive 4-week field study will be conducted in Barrow during March 1978. Chemical, physical, and optical properties of the aerosol will be determined. Daily aerosol samples and radiation measurements will be taken at the surface, coordinated wtih a near-daily, 3-week series of aircraft flights on which aerosol (and gas) samples will be taken and radiation will be measured. Goals of these flights include studies above and below the near-surface inversion layer, vertical profiles, tracking of crustal haze bands southward over the Pacific toward their Asian source, investigation of Prudhoe Bay as a source of Arctic pollution, and documenting aerosol concentrations far north of Barrow. Brief experiments on diffusion of vanadium out of the tundra and abundance of particles in pack-ice cores will also be attempted.

A preliminary series of aircraft-based radiation measurement will be made in the Norwegian Arctic to determine the vertical profiles of aerosol

there. Vertical profiles of Arctic aerosol are important for climatic calculations, but to date have only been measured at Barrow.

Extensive meteorological analysis will continue, both for specific field experiments and for general transport of pollution aerosol to the Arctic. Chemical analysis of several desert soils as a function of particle size, now in progress, will be concluded, from which it is hoped that the crustal contribution to the atmospheric aerosol can be better understood.

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I. Progress Report: August 1976 - July 1977

A. Highlights of results

This year has been an extremely productive one for our Arctic haze project. Briefly stated, our research has:

1. Confirmed by comprehensive neutron activation analysis (supplementing the short irradiations reported last year) that the composition of Arctic haze bands is much more nearly crustal than is the normal northern aerosol (Figure 11 of Section ID). Many elements which are highly enriched relative to the crust in normal northern aerosol are still somewhat enriched in Arctic haze bnads but vanadium (V), which normally is only moderately enriched, was nonenriched in the bands, i.e., the V/Al ratio was exactly crustal. From this we originally concluded and still believe that the haze aerosol of the bands of April and May 1976 was crustal (most likely desert dust) rather than pollution in origin. With a crustal V/Al ratio these bands could not have come from Europe or from the northeastern United States.

2. Further confirmed the crustal-dust nature of these bands by scanning electron microscopy for particle morphology and size and volume distributions; electron microprobe single-particle analysis for chemical composition. (Section ID, pp. 45-50)

3. Confirmed by detailed multi-level trajectory analysis (Section ID, pp. 16-22) that air from eastern Asia reached Barrow during the times that banded Arctic haze was seen there in April and May 1976.

4. Identified a major dust storm in eastern Asia at the proper time and place to have been the source of our haze bands. We followed this storm on the surface weather maps for nearly 3000 km over the Asian mainland,

Korea, and Japan, until it was out over the Pacific.

5. Identified the presence of easily detectable amounts of pollution aerosol in the surface atmosphere of Barrow, Alaska, primarily during the winter (Section ID, pp. 58-61). Much or most of this pollution is almost certainly derived from mid-latitudes via long-range transport, and is seen at Spitzbergen as well.

6. Posed a meteorological hypothesis to explain the large winter-summer differences in pollution aerosol concentrations in Barrow (and presumably the rest of the Arctic as well , Section ID, pp. 57-62; Section IE).

7. Broadened our concept of Arctic haze into two types. (Section ID, pp. 54-65). Type A refers to the banded, crustal haze from Asia, and Type B refers to the more diffuse, possibly pollution-derived, haze of the Arctic winter. The starting point for this idea was that the Asian dust bands did not increase the overall turbidity over Barrow when then they arrived in April 1976 (Section ID, p. 54).

8. Noted the similarity in winter pollution maxima and spring maxima of Arctic turbidity, and tentatively identified the unusually high Arctic turbidities with pollution aerosol, either directly or indirectly (Section ID, pp. 54-65). In spring (and probably also winter) Arctic turbidities appear to be significantly higher than subarctic turbidities, just the opposite of what one would expect.

9. Posed the question of whether pollution aerosol detected in the Arctic could be exerting a climatic effect, by warming the Arctic.

We have chosen to summarize our work for the first 18 months of this project by preprints of four of our publications - Sections IC through IF. The first of these, a brief introductory report written in December 1976 and

scheduled to appear in Nature about late August 1977, summarizes our initial results and conclusions. The second, and most comprehensive, article is a report on the entire project written for inclusion in the proceedings volume of the symposium "Desert Dust and Its Effects", held as part of the February 1977 Annual Meeting of the AAAS in Denver, Colorado. This article is essentially our entire progress report. The third and most recent publication has just been submitted to Science. It advances our hypothesis that pollution aerosol seems ubiquitous in the Arctic and has its transport from mid-latitude sources controlled by movements of the jet stream/polar front. Finally, the fourth article presents a summary of Dr. Schütz's Ph.D. thesis on long-range transport of Sahara dust westward over the North Atlantic. While not directly related to our Asian research, we include this article here for two reasons. (1) It was extensively expanded and rewritten after Dr. Schütz came to URI, based on discussions among him, Dr. Rahn, and numerous students and staff members of the GSO. (2) It indicates the type of work that would be done if we choose to concentrate our studies on the source of Asian dust. For example, we could try to determine the specific source areas, the times of the year and meteorological conditions for dust generation and the heights at which the dust is transported over the Pacific.

B. Publications

Publications supported wholly or partially by this contract include, in order of writing, the following articles:

Rahn, K.A., "The chemical composition of the atmospheric aerosol", University of Rhode Island Technical Report, 1 July 1976, 265 pp.

Rahn, K.A., L. Schütz, and R. Jaenicke, "The crustal component of background aerosols: its importance in interpreting heavy metal data", Proceedings of the World Meteorological Organization Technical Conference on Atmospheric Pollution Measurement Techniques (TECOMAP), Gothenburg, Sweden, 11-15 October 1976. (In press).

Rahn, K.A., R.D. Borys, and G.E. Shaw, "The Asian source of Arctic haze bands", Nature (~~in press~~) 268, 713-715.

Rahn, K.A., R.D. Borys, G.E. Shaw, L. Schütz, and R. Jaenicke, "Long-range impact of desert aerosol on atmospheric chemistry: two examples", Proceedings of the Saharan Dust Workshop, Gothenburg, Sweden, 25-28 April, 1977. Sponsored by the Ecological Research Committee (Secretariat for International Ecology, Sweden) of the Swedish National Research Council, Sveavägen 166, 15 tr, S-11346 Stockholm, Sweden (in press).

Rahn, K.A., R.D. Borys, and G.E. Shaw, "Asian desert dust over Alaska: Anatomy of an Arctic haze episode," Proceedings of the AAAS Symposium "Desert Dust: Origin, Characteristics, and Effect on Man", Denver, 22-23 February 1977, University of Arizona Press (~~submitted~~) (in press)

Schütz, L., R. Jaenicke, and H. Pietrek, "Saharan dust transport over the North Atlantic Ocean", Proceedings of the AAAS Symposium "Desert Dust: Origin, Characteristics, and Effect on Man", Denver, 22-23 February 1977, University of Arizona Press (submitted).

Rahn, K.A., R.D. Borys, G.E. Shaw, and S. Larssen, "Arctic-wide aerosol pollution", Science (submitted).

Graham, W., L. Schütz, and K.A. Rahn, "The transport of phosphorus to the North Atlantic by the Saharan dust plume", J. Geophys. Res. (to be submitted).

Papers presented at conferences but not yet published

Rahn, K.A., "Major research areas of atmospheric chemistry initiated by neutron activation analysis: Three examples". Presented at the Nuclear Applications Award Symposium: Nuclear Methods of Chemical Analysis, 173rd American Chemical Society National Meeting, New Orleans, Louisiana, March 21-25, 1977.

Rahn, K.A., R.D. Borys, and G.E. Shaw, "Air pollution in the Arctic - some emerging ideas", Presented at the conference "Sources and significance of natural and man-made aerosols in the Arctic", Norwegian Institute for Air Research, Lillestrøm, Norway, 27-28 April, 1977. To be submitted to J. Appl. Meteor.

C. Nature article: The Asian source of Arctic haze bands (12 pp.)

Rev

On the Asian source of Arctic haze Hazes

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Revised 4 April 1975

ABSTRACT

Arctic haze aerosol over Barrow, Alaska was collected on filters using a light aircraft. Its concentration, chemical composition, morphology, and trajectories to Barrow indicated that it had a strong source region in the great deserts of eastern Asia. The input of such large amounts of crustal aerosol into the Arctic was previously unknown.

"Arctic haze" refers to turbid layers of air which are found regularly over the pack ice north of Alaska during periods of clear weather[1]. These layers are diffuse, hundreds to thousands of kilometers wide, 1 to 3 km thick, and can occur as single or multiple bands of different heights at nearly any level in the troposphere. They are invisible from the ground, but may limit horizontal and slant visibility within a layer to as little as 3 to 8 km. Their color is grey-blue in the antisolar direction and reddish-brown in the solar direction, suggesting that they are true aerosol rather than ice crystals.

The initial, purely visual observations of Arctic haze were made more than 20 years ago. It was then forgotten about until 1972 when radiation measurements near Barrow, Alaska revealed unexpectedly high atmospheric turbidities, which were confirmed in 1973 and 1974[2]. The amomalous turbidity was partly found in distinct layers at altitudes of only a few kilometers. About 40%, however, was above 4 km, the effective ceiling of the aircraft used.

The 1974 observations raised the question of possible anthropogenic origin of the haze, because rough trajectory analysis for a persistent haze period during March and April 1974 suggested that the air had passed over the northeastern United States some days earlier. To check this possibility, we have used the chemical composition and morphology of the spring 1976 Arctic haze aerosol as indicators of its source, and concluded that it originated in the great Asian deserts. During April and May 1976 a series of 15 flights with a single-engine aircraft were made from the Naval Arctic Research Laboratory in Barrow, Alaska. Nine high-volume (90 to 360 m^3) samples of haze aerosol were collected on acid-washed 11-cm Whatman No. 41

cellulose filters. Nuclepore filter samples were taken concurrently for electron microscopy. Height of the haze layers was determined by a combination of visual observation, condensation-nucleus count, and turbidity profile.

The high-volume samples were subsequently analyzed at the University of Rhode Island for a number of elements with short-lived nuclides by neutron activation, with results shown in Table 1. The month-long sampling period had very low elemental concentrations at the beginning, a strong maximum in the middle by a sharp dropoff at the end. The high-concentration period coincided with visible haze bands, all of whose visual properties suggested Arctic haze. The bands did not come and go quickly; rather a single broad maximum of 6 to 13 days' duration was seen. During this period Al and Mn were increased in concentration relative to their initial and final values by over an order of magnitude, V by an order of magnitude, and Na and Ba by somewhat less than an order of magnitude.

The aerosol-crust enrichment factors (Table 1) for the elements,

$$EF_X(Al, \text{ rock}) = (X/Al)_{aerosol}/(X/Al)_{rock} ,$$

showed similar trends of high values in the early samples which decreased smoothly to unity or lower with increasing haze aerosol. For V the trend was most marked; indeed of these elements V has the highest enrichments in cities (5 to 500[3]). Taken together, the trends of these enrichment factors clearly indicated that the background aerosol was pollution-derived but that the haze aerosol itself was <u>crustal</u>, i.e. natural, just the opposite of what we had expected. Electron microscopy of the Nuclepore filters showed that angular crustal particles of diameter greater than roughly 0.4 to 0.8 μm are present in all samples, but with greatly increased

numbers during the haze episode. In contrast, most particles smaller than 0.4 to 0.8 μm diameter were nearly spherical; their abundances were much less haze-dependent. Although we have studies only this one episode, we feel that it may well be typical, as discussed below.

The source of the crustal haze aerosol must be very strong to account for its high concentrations. It seems to be the great Takla Makan and Gobi deserts of eastern Asia. Upper-level constant-pressure charts indicate that our haze period was preceded by several days of intense air flow from these deserts to Alaska. Figure 1 shows actual 700-mb isobaric trajectories for air arriving at Barrow before, during, and after the episode. Only episodic air had recently passed over the Asian deserts.

Several factors support the idea of an Asian source for Arctic haze. First, spring is the period of greatest dust storms in deserts. In our case, during nearly all of April 1976 the Asian deserts were filled with dust storms. Second, the composition of Arctic haze greatly resembles that of the Sahara aerosol (K.A. Rahn, L. Schütz, and R. Jaenicke, unpublished data). Third, flow patterns conducive to long-range transport from Asia to Alaska are strongest during the spring[4,5]. Fourth, the Asian deserts are farther north than the other deserts, lying mostly between 40° and 50° North as opposed to the more normal desert latitude of 20° to 30°. Fifth, the length of typical trajectories between Asia and Alaska (9000 to 12,000 km) is not out of line with the well-documented 6000-km path for transport of Sahara dust to Barbados.

An Asian source for Arctic haze explains or confirms many previous observations such as (1) Anomalously high turbidities over Barrow, especially during the spring[7] (Flow patterns between Asia and Alaska appear to be common from November through May); (2) The large size of the haze bands (see below); (3) Prior haze events at Barrow (For example, the episode of March-April 1974 was preceded by a few days of strong flow from the Asian deserts); (4) Prior measurements of anomalous turbidities over Fairbanks, Alaska (For example, 17-22 February 1976 was a whitish haze episode in Fairbanks which affected air at all levels and for which no explanation could be found. Visibilities were reduced from the normal 150 km to less than 30 km. Trajectory analysis now reveals that this air was also of Asian origin. Similar haze incidents with trajectories leading back to Asia seem to be a regular feature of the Fairbanks atmosphere during winter and spring; (5) The accumulation of brownish insoluble deposits in the pack ice north of Barrow, which under examination with a light microscope appear to be continental dust (R. Paquette, personal communication). The mineralogy and possible sources of this dust have also been discussed[8]. (6) Anomalous ice-nucleus concentrations at College, Alaska, Blue Glacier, Washington, and Nagoya, Japan during February and March 1968, for which trajectory analysis showed eastern Asia to be the probable source[9]; (7) A case of abnormally high condensation nucleus concentrations at Barrow in March 1970[10], which was explained as pollution from Prudhoe Bay to the east but which may have originated from Asia because the large-scale flow for this period was from those deserts.

The mass of desert dust transported into the Arctic seems to be very great. For the 5-day episode over Fairbanks discussed above, columnar mass loadings derived from radiation measurements showed values of about

45 mg m^{-2}(for aerosol density 2.5). For a point source in Asia and a dispersion angle of 5^{o}(typical of volcanic plumes), the dust cloud would be 900 km wide at Alaska, or about the width of the state itself. (Indeed during this incident visibility was poor over the entire north half of the state). Such a plume travelling at 80 km hr^{-1} would carry 4000 tons of dust into the Arctic per hour, or a half-million tons over the 5-day episode. This is equivalent to skimming off a 0.2-μm layer from a $10^{o}x10^{o}$ desert.

Clearly, more detailed studies are needed to refine our ideas about this significant phenomenon. Also, possible climatic effects of such large amounts of aerosol in the Arctic should not be overlooked.

ACKNOWLEDGEMENTS

This work was supported by Office of Naval Research contract N00014-76-C-0435. The cooperation of many staff members of the Naval Arctic Research Laboratory, Barrow, Alaska is gratefully acknowledged. Samples were chemically analyzed using facilities of the Rhode Island Nuclear Science Center, Narragansett, Rhode Island.

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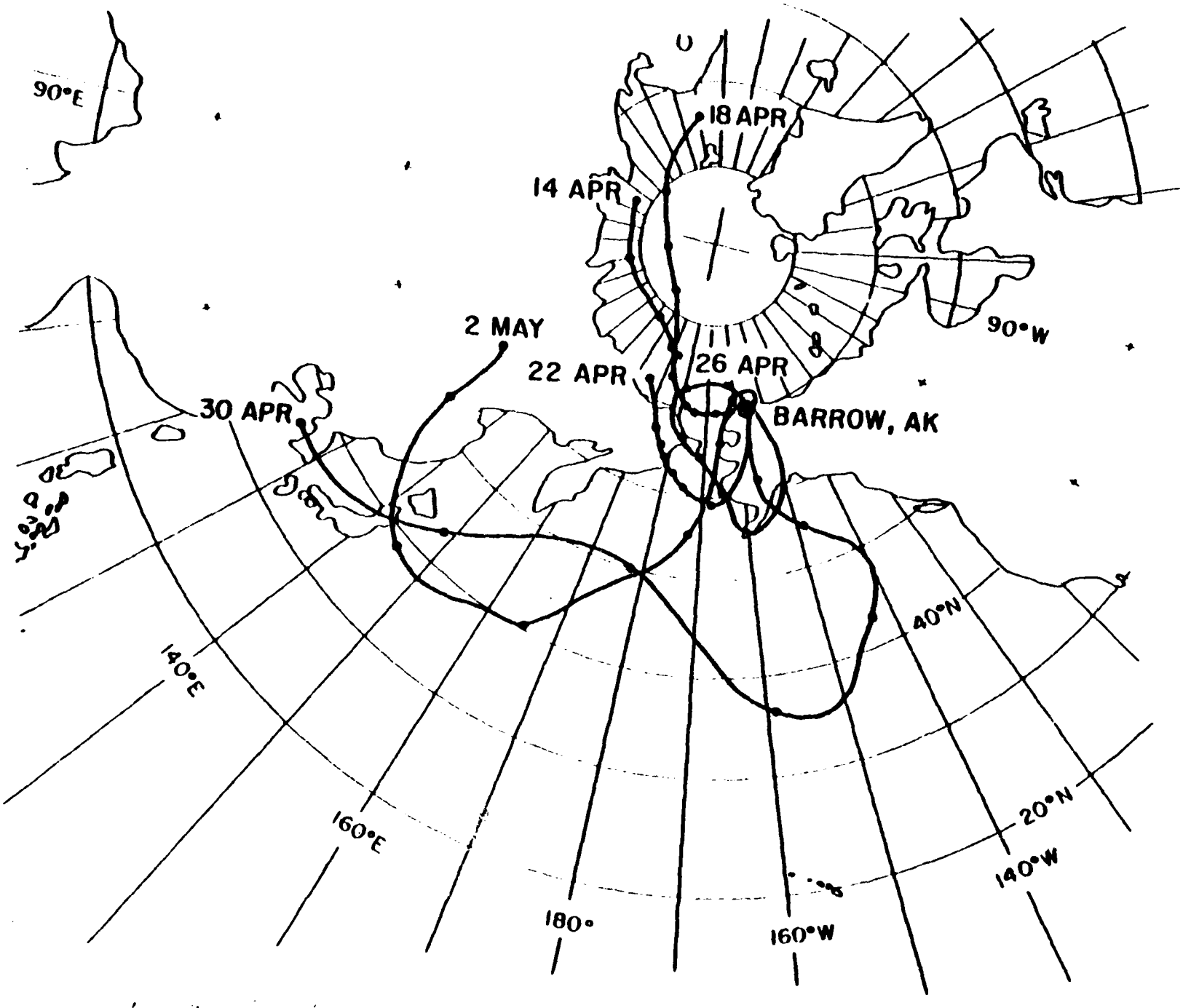
TABLE I.

ELEMENTAL CONCENTRATIONS AND ENRICHMENT FACTORS FOR AIRCRAFT FILTER SAMPLES

SAMPLE:	AB	CD	E	F	G	IJ	KL	M	N
				Concentration, ngm^{-3} (ambient)					
Al	10.0 ± 0.9	17.0 ± 1.0	35 ± 2	34 ± 2	91 ± 5	240 ± 10	203 ± 10	240 ± 10	14.7 ± 1.7
V	0.035 ± 0.006	0.148 ± 0.010	0.126 ± 0.012	0.094 ± 0.010	0.22 ± 0.02	0.40 ± 0.03	0.37 ± 0.02	0.40 ± 0.04	0.0192 ± 0.0100
Mn	0.146 ± 0.012	0.36 ± 0.02	0.59 ± 0.04	1.58 ± 0.03	1.55 ± 0.08	3.0 ± 0.2	3.1 ± 0.2	3.4 ± 0.2	0.21 ± 0.02
Na	12 ± 10	32 ± 11	<30	21 ± 16	47 ± 27	58 ± 15	37 ± 11	37 ± 30	<20
Ba	0.4 ± 0.2	0.4 ± 0.2	<0.8	<0.5	1.6 ± 0.6	2.0 ± 0.2	2.0 ± 0.2	1.9 ± 0.5	<0.5
				Enrichment Factor, (Al, rock)					
Al	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
V	2.1	5.2	2.2	1.66	1.46	1.00	1.10	1.00	0.79
Mn	1.25	1.81	1.44	1.46	1.46	1.07	1.31	1.21	1.22
Na	3.4	5.4	<2.5	1.77	1.48	0.69	0.52	0.44	<4
Ba	8	5	<4	<3	3.5	1.7	1.8	1.5	<6
DATE	April	14,15 April	17 April	19 April	21 April	28,29 April	30 April, 1 May	3 May	5 May
VOLUME, m^3	249	341	149	176	110	309	365	88.7	108
ALTITUDE km	3.3	2.0, 1.2	2.1	2.1	3.0	1.8, 2.3	2.1, 2.0	2.5 to 2.8	1.8

FIGURE CAPTIONS

Figure 1. 700-mb isobaric trajectories of air to Barrow. Numbers refer to date of arrival at Barrow. Solid circles each 24 hours along trajectory.


90°E
18 APR
14 APR
2 MAY
90°W
22 APR
26 APR
30 APR
BARROW, AK
40°N
140°E
160°E
180°
160°W
20°N
140°W

D. AAAS article: Asian desert dust over Alaska: Anatomy of an Arctic haze episode (108 pp.)

Asian desert dust over Alaska: Anatomy of an Arctic haze episode

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3. The turbidity problem

ABSTRACT

A several-day series of Arctic haze bands over Barrow, Alaska in April and May, 1976, has been found to be crustal dust from the Gobi Desert, Loess Plateau, and possibly also the Takla Makan Desert of eastern Asia. The haze aerosol which formed the bands was sampled *in situ* from a light aircraft. Elemental analysis of the particles by neutron activation revealed that they were crustal or near-crustal in composition, and in particular that they could not have originated in Europe or the northeast United States. Electron microscopy showed that the haze particles had a mass-median radius of nearly 2 μm, which meant that they had probably travelled more than 5000 km from their origin. They were usually angular in shape, as would be expected for crustal fragments. Several particles had a platy structure characteristic of micas. Single-particle analysis with an electron microprobe showed that most of the larger particles were rich in silicon and aluminum, two characteristic crustal elements.

Trajectory analysis showed that the haze-containing air had passed over the arid and semi-arid regions of eastern Asia a few days earlier. During all of April and May, 1976, these regions had frequent dust storms, one of the strongest of which occurred seven days before the dust was first observed over Barrow, and could be traced from surface meteorological observations nearly 3000 km from the Gobi Desert south to the Loess Plateau, then northeast to Korea and Japan. This storm may have contributed a substantial fraction of the total dust observed over Alaska.

Turbidity measurements taken at Barrow before, during, and after the episode revealed that the Asian dust layers did not increase the total turbidity through a vertical column - rather, the turbidity actually decreased somewhat during the episode and sharply thereafter. Meteorological analysis has shown that this decrease was due to a massive influx of southern air, that is, air south of the jet stream, over Barrow as the jet stream temporarily migrated nearly to the North Pole. The hypothesis is advanced that the high prehaze turbidities which independent evidence has shown to be characteristic of Arctic air masses during winter, are pollution-derived with Europe and the northeast United States as the most probable sources. If this is the case, then distant natural and anthropogenic aerosol sources may be effectively altering the radiation balance of the Arctic.

I. Introduction

At the present time there is a generally increasing awareness of the major contribution that deserts make to the earth's atmospheric aerosol burden. The Sahara is by far the best-studied region in this regard. Of its annual dust output, about 260 million tons are transported over the North Atlantic Ocean in the direction of the Caribbean. About 30 million tons remain airborne as far as Barbados, a distance of 5000 km (Schütz, 1977; Schütz et al., 1978)

A quick glance at a climatological map or a globe is enough to reveal that the deserts of eastern Asia also ought to be very strong sources of atmospheric dust, much of which would be expected to travel long distances through the atmosphere in the same way that Sahara dust does. The arid and semi-arid regions of eastern Asia have long been recognized for their highly mobile dust. Dust storms in the Gobi Desert of China and Mongolia are legendary in their fury (Hedin, 1968, for example). Lesser known but major sources of dust are the Takla Makan Desert of extreme western China and the Loess Plateau of northwestern China (Figure 1).

A search of the literature, however, shows that very little is known about the Asian deserts as dust sources. A number of pieces of indirect evidence can be found which attest to dust transport from Asian deserts over the Pacific Ocean--the areawide distribution patterns of illite in deep-sea sediments (Griffin et al., 1968); the presence of quartz in Hawaiian soils and Pacific pelagic sediments (Rex et al., 1969; Syers et al., 1969; Clayton et al., 1972), where it could not have been formed locally; and the apparent transport of ice nuclei (clay-mineral

Figure 1. Asian base map, showing the Gobi Desert, the Takla Makan Desert, and the Loess Plateau.

particles) from Asia to North America (Isono et al., 1971). Yet no direct evidence about the origin and frequency of Asian dust storms, the annual production of dust, its transport, or its fate seems to be available.

The writers stumbled onto Asian desert dust in the atmosphere quite by accident. During an investigation of Arctic haze over Alaska in the spring of 1976, the strong haze bands seen there were found to be Asian desert dust. Using chemical, physical, optical, and meteorological techniques, the history of this dust episode has been reconstructed in some detail--from the time and place of its generation until its observation over Arctic Alaska several days and 12,000 to 15,000 km later. This paper describes the anatomy of this event as a case history of a previously unrecognized but apparently common phenomenon.

II. Generation of the dust

The winter half-year, roughly November through May, is the time of great Asian dust storms (Cressey, 1934). During this period the soils of the arid and semi-arid Asian source regions are at their driest, in response to the cold, dry prevailing winds from the north. Furthermore, the circulation over northwestern China is much more vigorous during winter than at other times of the year. By contrast, the summer season is marked by moister air from the south and less intense circulation, which combine to greatly impede the formation of dust storms.

April and May 1976 were no exception to this pattern. Both months had one or more active dust storms or suspended dust clouds over

eastern Asia at all times (April had more storms). The life histories of these dust events reveal a very complex pattern of behavior, wherein no two storms were quite alike. Many of the dust storms were short-lived, disappearing after about 12 to 24 hours. Some storms appeared to be quenched by surrounding bands of precipitation; in other cases there was no apparent reason for their disappearance. Some dust storms lived on for several days after their generation, and could be followed on the surface meteorological maps as broad or narrow plumes. All the dust storms probably contributed in greater or lesser measure to a quasi-steady supply of dust to the air aloft, which could then travel rapidly over great distances.

This section examines in some detail the surface dust records for eastern Asia between 20 and 27 April 1976, the period of generation of the dust that was observed a week later (29 April through 4 May) as banded Arctic haze over Alaska. There were six major dust storms during this period, four of which were lost track of at the surface and two of which could be followed for nearly 3000 km. One of these two storms was clearly the largest of the period and may have accounted for much of the aerosol observed in Alaska; we pay special attention to it here.

Figures 2 and 3 are pictorial representations of the surface meteorological conditions over Asia at 12-hour intervals from 0Z (0 GMT) of 20 April through 12Z of 25 April 1976. High- and low-pressure areas, associated fronts, and major directions of air flow are shown. In addition, regions of precipitation are indicated by sketched-in "clouds." Most importantly, stations reporting dust in the air, either actively

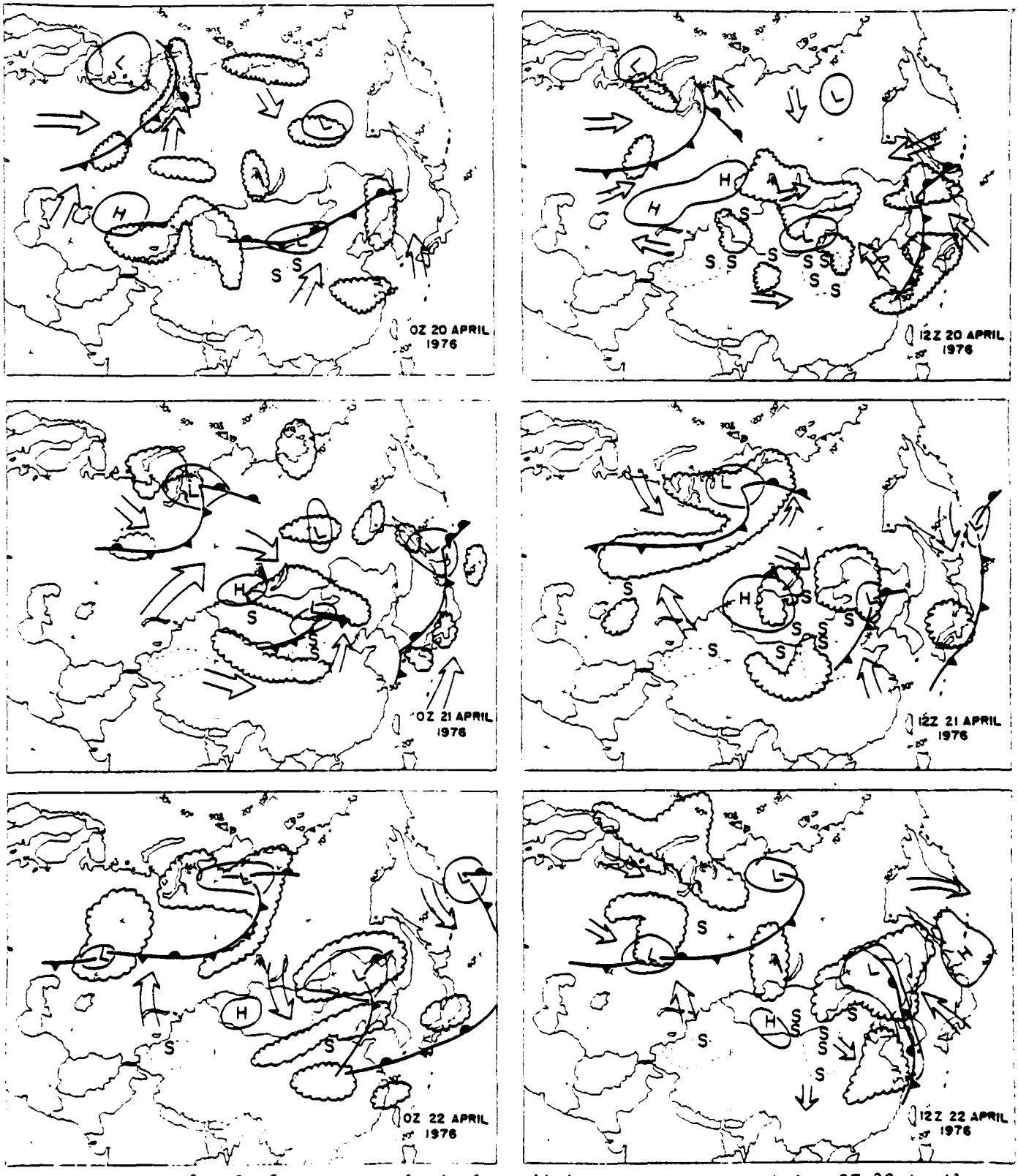


Figure 2. Surface meteorological conditions over eastern Asia, 0Z 20 April through 12Z 22 April 1976. Arrows show air flow, "S" represents dust in the air, and the "clouds" symbolize zones of precipitation.

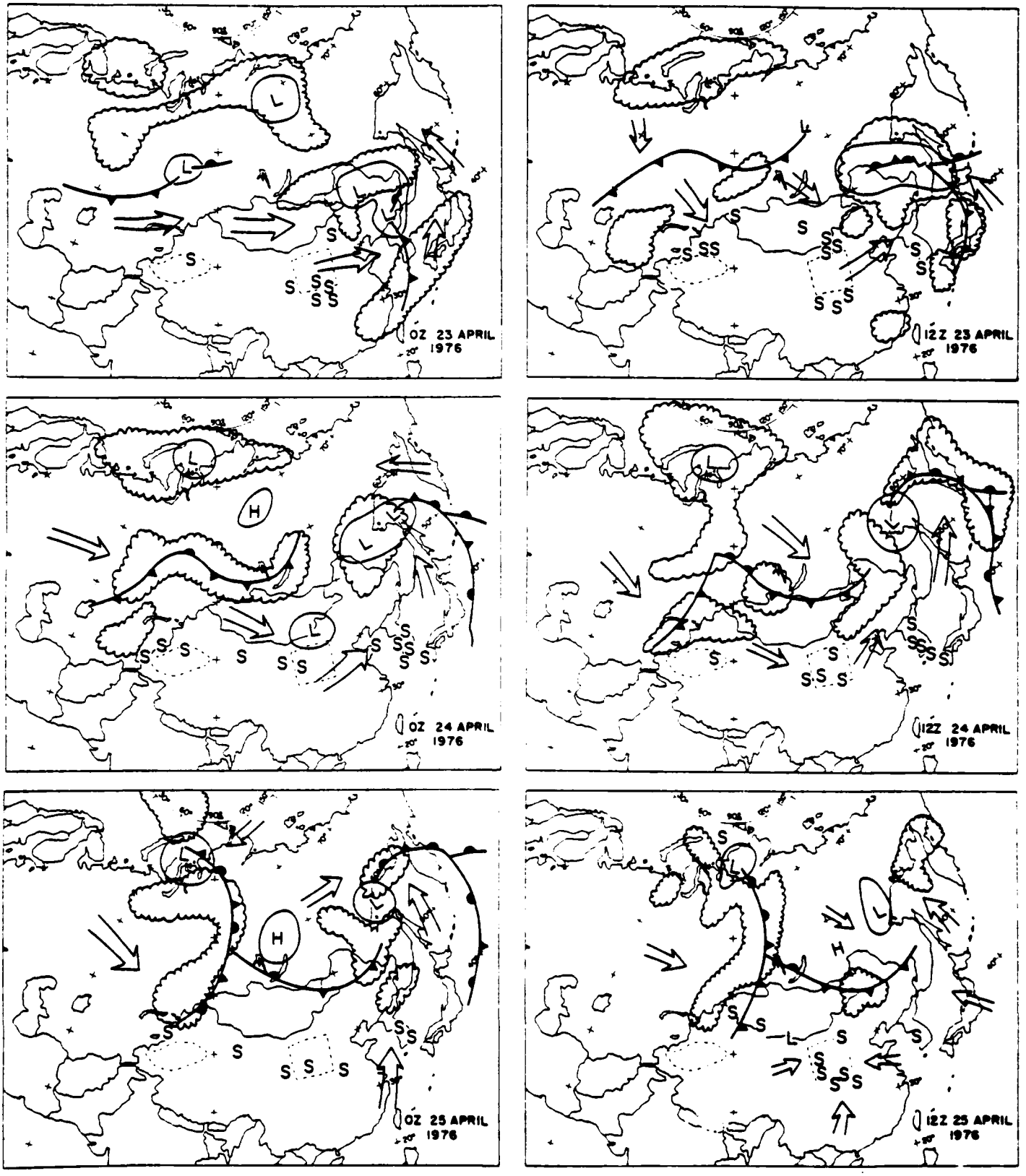


Figure 3. Surface meteorological conditions over eastern Asiz, 0Z 23 April through 12Z 25 April 1976. Arrows show air flow, "S" represents dust in the air, and the "clouds" symbolize zones of precipitation.

being generated or suspended from previous generation, are shown by the symbol S.

At 0Z of 20 April 1976 an east-west-elongated low-pressure area with an associated stationary front was situated over northern China and Mongolia. To the northwest of this front, in western China and Siberia, lay cold dry Arctic air; to the southeast, in China, lay warm moist tropical air. Very light dust activity was reported for the area, the only two dust clouds occurring in the Loess Plateau near the southern margin of the dry air. By 6Z of the 20th the eastern end of this low had spawned a second, less intense low over eastern Manchuria, which took the frontal system with it and subsequently moved northeastward out over the Pacific Ocean, leaving the original low behind over central Mongolia with no fronts. During the next 12 hours this original low moved slightly to the southeast and began to fill, i.e., weaken. To the west, however, a large high-pressure center, which formed the southernmost extent of a long ridge of high pressure stretching southwards from nearly the North Pole, built even more strongly, so that the pressure gradient between the low and the high showed a net increase. As a result, strong northerly air flow was established in the region between these two centers. At 12Z of the 20th active dust generation was reported in the Gobi and Takla Makan Deserts, as well as in the Loess Plateau. Note that the dust, while surrounded by zones of precipitation, was always found outside these regions. In no case was a dust storm or cloud reported inside a region of active precipitation, presumably because the moist ground did not emit dust and the rain or snow efficiently scavenged pre-existing dust from the air. By 18Z of the 20th

a front had appeared across the low, with the most intense circulation behind, or to the north of the cold-front (westward) side. This front first appeared on our maps for 0Z of the 21st, at which time the dust activity had quieted back down to three stations reporting. By that evening, however, there was again much active dust generation in the Gobi Desert. In the region of intense flow between the high and the low centers, six stations reported dust. This dust was surrounded by three large zones of precipitation, however, and by the following morning, 0Z of the 22nd, it had practically disappeared.

(Here one may note the apparent existence of a marked diurnal variation in dust frequency for this region. Much more dust was consistently reported in the air at 12Z [approximately 1900 local time] than at 0Z [approximately 0700 local time]. This was perhaps because the daytime insolation both dried out the soil and produced gustiness, which raised clouds of dust in the late morning and afternoon. As the winds died down during the evening and the ground became moister from the increased relative humidity, dust generation ceased and the cloud either settled out or was scavenged by precipitation. It is interesting that this pattern persisted through 0Z of the 22nd, during which time the pressure gradient over the Gobi Desert and, consequently, the average surface winds were increasing.)

During the next 24 hours a stronger and more concentrated dust storm was generated that did persist, though, and could be tracked for at least 72 hours and nearly 3000 km at the surface. We will now examine this period in some detail.

The dust storm began during the daylight hours of the 22nd of

April in the Gobi Desert. By that evening (12Z) it appeared fully developed and had shifted southward, with six stations of the Gobi Desert and the Loess Plateau reporting dust. Of the dust reported over the Loess Plateau, some appeared to have been transported from the Gobi and some originated locally. At first glance this storm greatly resembled the one 24 hours earlier, but there were several important differences which gave the storms entirely different life histories. The area of dust generation was now both the Gobi Desert and the Loess Plateau, whereas the previous day's dust had been limited to the Gobi. Prevailing winds were now northwesterly, as opposed to their more northerly orientation 12 to 24 hours earlier. More importantly, though, the air mass in which the dust was being generated was free of precipitation and much drier than before. Warming of the cold Siberian air mass over these more southerly regions had raised the temperature to typically 20 to 30°C above the dewpoint, corresponding to relative humidities of 10 to 20%. Although the winds of this period were not stronger than before, the extreme dryness allowed more dust to be generated and less to be removed. By the following morning, (0Z of the 23rd) in contrast to previous mornings, this dust still remained in the air, with five stations of the lower Loess Plateau reporting dust. Surface winds had now shifted to the west.

The low-pressure area was now moving rapidly to the northeast, with this dry, dust-laden air mass trailing behind. By the evening of the 23rd (12Z) surface winds had shifted to the west-southwest. The dust cloud had evidently passed still further south and then turned to the east, because there were three stations at the extreme south of the

Loess Plateau reporting dust (none in the center), as well as one in Korea and one at Cheju Island, just south of Korea. The wind speeds and directions over this period were such that these latter two locations, even though they were more than 2000 km from the Loess Plateau, could have been part of the dust load transported from there, rather than representing locally generated dust.

Twelve hours later, the following morning (0Z of the 24th), in response to the continuing northeastward motion of the low center and its fronts, the dust cloud had also spread more to the northeast. Whereas the previous two dust reports from the Korean vicinity at 12Z of the 23rd had apparently represented only the narrow leading edge of the advancing cloud, this report showed the fully-developed center and edges of the cloud. There were now six sites in the area of Korea and Japan reporting dust: three from Korea itself, plus Cheju Island, Nagasaki, Japan, and the tip of the Chinese mainland near Weihai. Weihai and Nagasaki extended the boundaries of the observed dust cloud to the north and south, respectively, and the site on the eastern coast of Korea extended its leading edge. Again, the direction of apparent motion of the dust cloud very nearly paralleled the motion of the front and the direction of the surface winds. The cloud had apparently completely left its area of origin on the Chinese mainland, because there were no more stations in the southern Loess Plateau reporting dust.

By the following evening (12Z of the 24th) the dust cloud had advanced significantly farther to the northeast. The trailing edge was apparently out over the Yellow Sea, because Weihai was dust-free. Also, no dust was reported at Cheju Island. A new station, at Hamhŭng, Korea,

showed the farthest-north advance of the dust to date.

By the following morning (OZ of the 25th) the dust cloud had mostly passed Korea, because only two locations there still reported dust. The flow at this time was nearly to the north, so that the main dust cloud was probably between Japan and the Asian mainland. Twelve hours later only one site reported dust, and after 12 more hours (not shown in Figure 3) Korea reported no more dust.

Figure 4 shows a summary of the positions of the dust cloud from the 22nd through the 25th of April, 1976. The original motion southward, followed by curvature to the northeast, is clearly seen here. This path is virtually identical to the upper-level air flow over this region. On the 500-mb contour maps (Figures 22 through 24), for example, where the flows are simpler and easier to trace than from the surface maps, a cyclonic wave is seen to pass over the Gobi Desert and Loess Plateau on the 22nd and 23rd of April. With the passage of the wave the flow progressed from northerly to westerly, and as the dust advanced it quickly came into a region of near-southerly flow.

At this point observations of the dust cloud ceased, until approximately OZ of the 29th, when it was again observed over Barrow, Alaska. By constructing long-range trajectories for the intervening period we have been able to satisfactorily trace where the dust must have gone, and demonstrate that what was seen over Alaska was indeed the same cloud that originated over Asia. The discussion in Section III shows this reconstruction.

It was earlier stated that two of the six dust storms between 20 and 27 April 1976 could be followed from surface records for more than

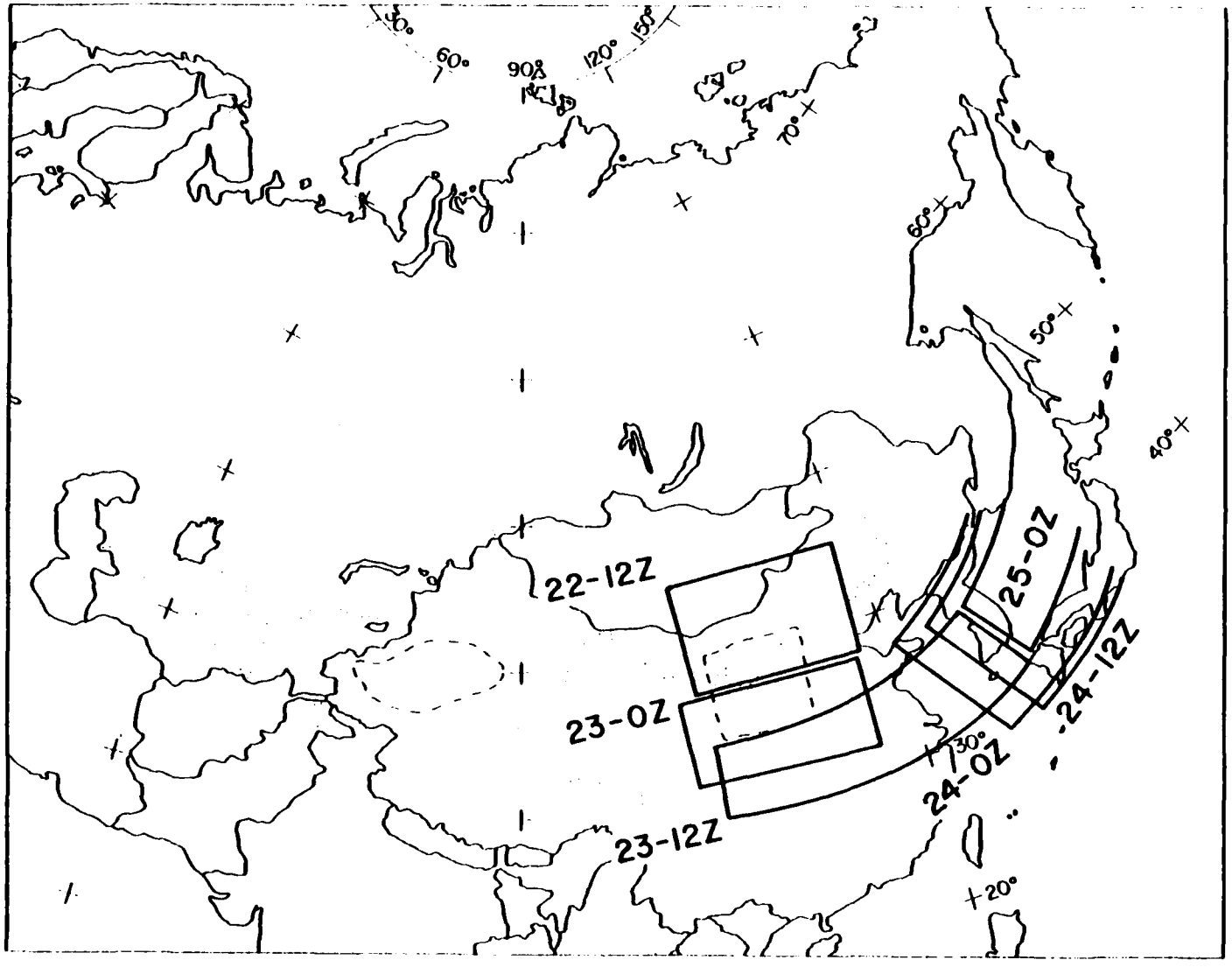


Figure 4. Summary of positions of the dust cloud for the persistent storm of 12Z 22 April 1976.

2000 km. Of these we have discussed only the first and larger one so far. The second, considerably smaller in scale and different in characteristics, is interesting in its own right. It is first clearly seen as four dust reports from the Loess Plateau on 12Z of the 14th. These reports may have been related to some scattered dust reports to the northwest 12 and 24 hours earlier; the latter were generally upwind of the former and at about the right distances for the dust to have been advected from as far away as the Takla Makan Desert. Indeed, the presence of five dust reports generally upwind of the Loess Plateau on the morning of the 24th (0Z) suggests that another major dust storm may have been generated the day before which did not disappear the following night. At any rate, this storm, if it is to be identified with the previous reports, seemed to be very long and narrow compared with the previously discussed broad storm and persisted through 0Z and 12Z of the 25th while the first storm was moving northeastward out to sea over Japan and Korea. By 0Z of the 26th (not shown here) it had completely disappeared at the surface, which is similar to the partial disappearance of the first storm over the eastern mainland just before it was detected in Korea. At least, the meteorological stations in eastern China seem not to report these storms very often as they pass; perhaps because they are lifted up over the warmer, moister air of the coastal lowlands and require a certain travel time before their giant particles can fall to the surface. On 12Z of the 26th Nagasaki again reported dust in the air, which was followed after 12 hours by an additional report of dust from a ship about 10°, or nearly 1,000 km, due east of Nagasaki. The upper-level wind patterns at this time suggest that the

dust over the Loess Plateau, Nagasaki, and the ship was part of one and the same cloud, in this case a very long and narrow dust plume tracked nearly 3000 km from its source, and possibly 5000 km if the Takla Makan had been the true source.

At this point it may be appropriate to comment on the unexpected locations and mechanisms of origin of the Asian dust. The common picture of large-scale dust generation to the atmosphere is that it takes place in the immediate vicinity of strong fronts, where sufficiently powerful and gusty winds are available in combination with broad-scale uplifting to insure that dust, once generated, can be raised to heights of a few kilometers where it can be carried away by the upper-level winds. Cold fronts, because of their intensity and slope, are usually considered the prime agent for creating large dust storms. For example, Danielsen cites the case (in Jackson et al., 1973) of a large dust storm on 22 April 1963 in Arizona and New Mexico where the dust was generated just ahead of an advancing cold front, where the greatest winds of the area were found. He states specifically that the principal arid regions of the globe, which lie outside of the region between the Tropics of Cancer and Capricorn, are vulnerable to this type of dust storm. In this regard, then, it is interesting that the dust storms of April 1976 in China and Mongolia, while outwardly satisfying Danielsen's criteria for the type of origin, seem instead to have very little association with cold fronts. For example, on 0Z of 20 April there was some dust generated well ahead of the stationary front, on 12Z of the same day there was a great deal of dust in the same vicinity but the front had disappeared, and on 0Z of the 21st there was dust both ahead of and

and behind the newly formed cold front in the Gobi Desert. After this time the front advanced to the south and east, but the area of dust generation remained in the Gobi Desert and the Loess Plateau. From 12Z of the 22nd through 12Z of the 25th of April, the dust generation was far removed from all fronts and essentially decoupled from them. If anything, the preferred area of dust generation seemed to be well behind the advancing cold front. Careful review of the surface meteorological maps for the period suggested that the dominant influence in dust generation was the presence of dry air from the north rather than wind speed as such. Apparently when these Asian soils are moist they cannot become airborne, but after a day or so of drying out under strong dry winds and insolation only relatively low wind speeds are needed to create dust storms. This reasoning is mostly conjecture, however; detailed understanding of the important processes in the generation of Asian dust needs a great deal of further study.

There also seems to be some disagreement about the source region of Asian dust storms. Cressey (1934, p. 75) remarked that the Gobi Desert was only an indirect source of dust observed downwind from it: *"It is popularly supposed that the dust storms (of the North China Plain and the Loess Highlands) originate in the Gobi Desert, but such is only indirectly the case. The Plateau of Mongolia is today relatively free of fine material, its fine weathered products having long since been blown southward to build up the thick deposits of loess. These loess hills together with the dry flood plains of the rivers and the bare fields are the immediate source of most of the dust of North China. Each strong wind lifts fine material and carries it another step on its*

southward journey, so that the dust in any particular storm is chiefly from local sources."

Cressey's hypothesis can be checked in a straightforward manner. Gillette et al. (1972) and Schütz and Jaenicke (1974) have found eroding soils from Nebraska and the Sahara Desert, respectively, whose mass distributions suggest that their fine particles have been blown out in the way that Cressey suggests for the Gobi. Similar analysis for properly chosen regions of the Gobi Desert, together with dust-storm records for these regions, should confirm or deny Cressey's idea. This subject is of more than mere academic interest. Violent dust storms rage through the Gobi (Hedin, 1968). Should these storms be proven to be only local in effect, study of the source regions for Asian dust which is transported long distances in the atmosphere would have to shift more to the Loess Plateau. In this connection it is interesting to note that there is no general agreement about which types of regions within the Sahara Desert are the true source regions for its dust (SIES Report, 1978). For example, some researchers feel that the sand dunes are source areas, but this is denied by other researchers who note that the dunes are "blown out" and have too few fine particles to be effective sources.

III. Transport to Alaska

A good feeling for the track of the dust cloud over the Pacific Ocean can be obtained from the general synoptic situation for the period of 21 April to 8 May, maps of which are shown as Figures 21 through 23. The dust advanced to the east and northeast along the jet to approximately the longitude of the Bering Strait, at which point it became associated with the strong Aleutian low and curved northward, passing

over Alaska and the Arctic Ocean. Details of this synoptic situation differed from day to day; the most persistent synoptic feature was the Aleutian low, which provided the basic northward impetus to move the dust to Alaska.

A more accurate idea of the actual paths taken by the dust on its way to Alaska can be obtained by air-mass trajectory analysis. At the outset of this discussion it must be clearly stated that all forms of trajectory analysis, like weather forecasting, become progressively uncertain with increasing distances or times. For the conditions of our calculation-- transport times of several days and distances of approximately 12,000 to 15,000 km --rather great uncertainties must be expected, and the resultant trajectories must be considered as indications rather than fact.

Isentropic trajectories, in which vertical motions of the air parcels under consideration are taken into account, are best. This type of trajectory is very difficult and time-consuming to calculate, however, and is seldom used. More commonly, one simply follows the air parcel along a constant-pressure surface, usually either 500 mb (approximately 5500 m altitude) or 700 mb (approximately 3000 m). Isobaric trajectories are easier to calculate than isentropic trajectories, but are more uncertain. The latter are commonly used, however, with good success to indicate the general area of origin of an air mass. Isentropic trajectories were beyond the scope of this project. We did attempt to make a compromise, however, between isentropic and isobaric trajectories by constructing multi-level isobaric trajectories, as we discuss below.

Forward trajectories were constructed for various Asian source areas for 21 through 26 April 1976, with all starting times 0Z. Source areas were based on the locations of observed dust storms, and were made rectangular. They corresponded roughly to the Gobi Desert, the Gobi Desert plus the Loess Plateau or the Loess Plateau alone. The first source areas were in the Gobi Desert, then were moved progressively southward from the 21st through the 26th of April. From each corner of the rectangle an individual trajectory was constructed. The first 12 hours of each trajectory were done at 850 mb (approximately 1500 m), the second 12 hours at 700 mb, and the rest of the trajectory in most cases was done at 500 mb. This corresponded to a vertical velocity of 5 cm s^{-1} during the first 24 hours, which is comparable to that observed in large-scale cyclonic motion. (The first part of all trajectories was indeed cyclonic, because of the presence of the upper-level trough over the source areas.) Whenever anticyclonic motion persisted for more than 12 hours at the 500-mb surface, the trajectory was dropped to the 700-mb level, and later returned to 500 mb when cyclonic motion persisted. Anticyclonic motion was most often found over the middle of the Pacific Ocean and over Alaska.

These area trajectories are shown in Figure 5, as six individual maps. The original rectangle, chosen according to locations of dust storms for that entire day, are shown, plus successive daily positions of the advected, distorted air masses. Corners are labelled A through D, with subscripts denoting the number of days of travel. For April 21st all four corners of the square had slow trajectories which remained well to the south of Barrow even after six days. Point B took the most northerly

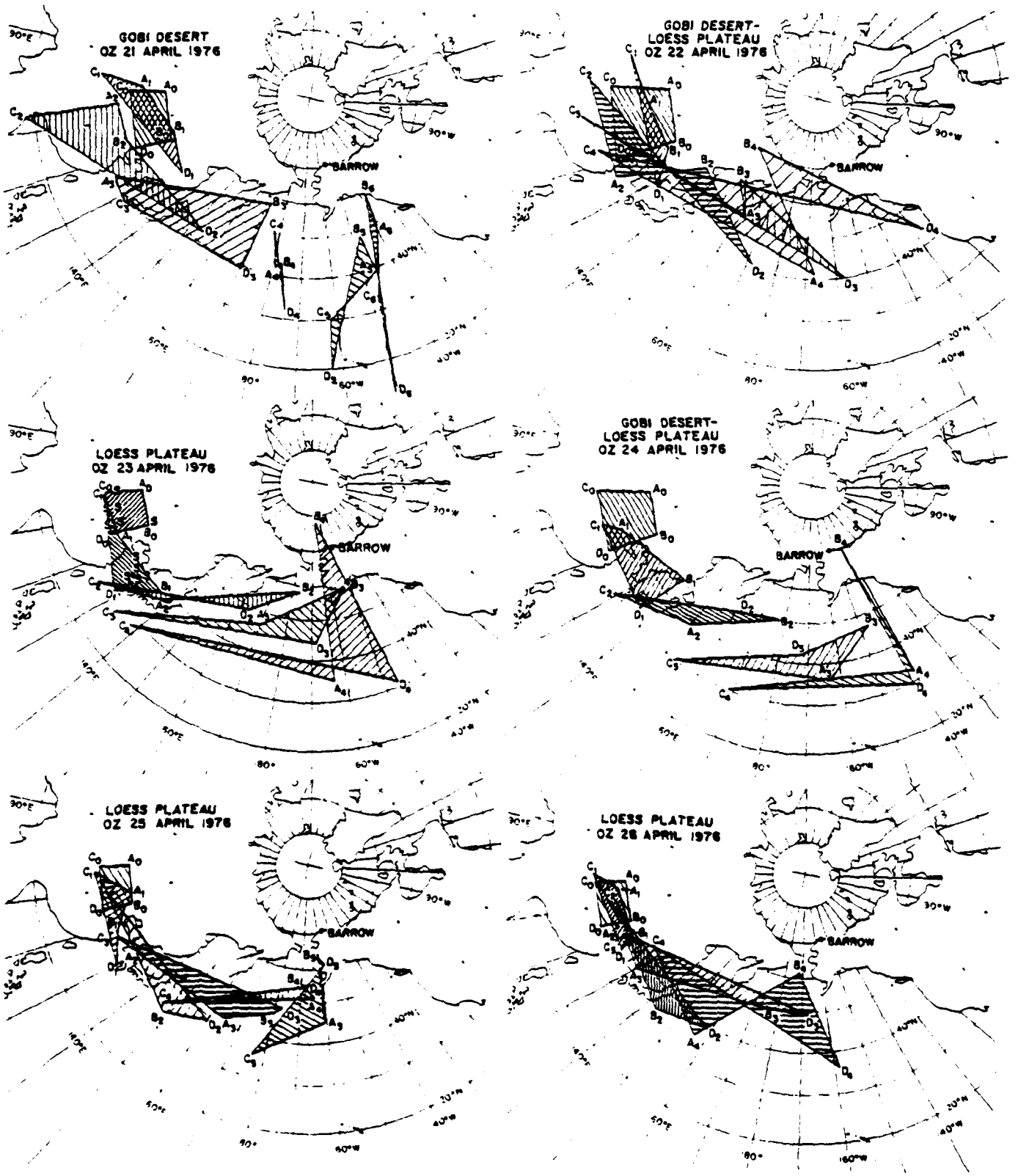


Figure 5. Forward mixed-level trajectories from Asia, 21 through 26 April 1976. Initial rectangles are locations of dust for that data; subscripts signify the number of days of travel.

course of the four and touched southeastern Alaska after six days, but its further trajectory was to the northeast and did not approach Barrow. Points A and C remained at lower latitudes and were also heading northeastward after the sixth day. Interestingly, point D passed south of Hawaii, which shows that under certain conditions Asian dust should reach those islands. This supports the idea of an Asian source for quartz particles in Hawaiian soils, whose presence has been noted earlier (Syers et al., 1969; Rex et al., 1969).

The trajectories for 22 April 1976 were considerably faster than those for the 21st because the center of the upper-level jet was now over the source area, whereas one day earlier it lay well to the northwest. Also, as the wave advanced it strengthened, which helped increase wind speeds on the 22nd. Thus the plume reached Alaska in less than four days, with a point roughly midway between B and D of the original square heading directly for Barrow. The plume was, however, very spread out, because point C moved so slowly that it remained over the Chinese mainland through the fourth day.

By 0Z of the 23rd the transport conditions were ideal to carry Asian dust to Alaska. On this day the upper-level winds had the best combination of intensity and orientation to transport dust from the Loess Plateau to the northeast. Indeed, of the six days considered here the trajectories did actually move the fastest on this day. Point B reached Barrow in just over 3-1/2 days, a remarkable speed. The 23rd is particularly important because it had the greatest number of stations reporting dust of any day during the period of dust generation. The major dust storm which could be followed from surface observations across

China and out over Korea and Japan is best represented by the trajectories of this day. For this reason the 23rd is discussed here in more detail than the others. After the first day the rectangle had already been severely distorted, with point B moving the fastest, points A and D somewhat more slowly, and point C the slowest. The six dust reports for 0Z of the 24th are also shown within this area. Note the very close correspondence between their locations and the predicted location of the dust plume. Throughout days 2, 3, and 4 point B continued to move the fastest, point C the slowest, and points A and D at intermediate speeds. After about 3-1/2 days of travel point B had reached Barrow, having travelled northeastward to the vicinity of Anchorage, then northward to Barrow and beyond. Points A and D, however, stayed moderately south of A throughout the first two days, then veered far to the south around the Aleutian low, after which they probably never reached Alaska. They may have reached North America somewhere between 40 and 50°N latitude. Point C, the slowest, had not even moved beyond the longitude of Japan after four days.

This figure illustrates graphically some of the interesting aspects of long-range dust transport. First, it shows that aerosol from the area of Asian dust storms can quite easily reach Barrow, Alaska in a few days. Second, it shows just how different the tracks of aerosol from geographically adjacent areas can be after only three to four days. Finally, it illustrates how a small "blob" of aerosol can be quickly stretched out into a long thin plume when wind speeds across the original blob are significantly different. Admittedly this case may be a bit extreme because the source area lay just under the strongest region of the

upper-level jet, where the horizontal wind shear was unusually great. On the other hand, Rossby (1959) suggested that such stretching or distortion may be common for the atmosphere. He showed a typical example of a square source area becoming greatly deformed after only 24 hours.

A similar area trajectory for the 24th of April showed a nearly identical result, but with slightly slower travel speeds. Point B arrived at the latitude of Barrow after four days instead of 3-1/2, and was displaced somewhat to the east of the plume from the 23rd, but the overall picture was very much the same.

Trajectories for the 25th were considerably slower. After more than five days points B and D appeared to pass over Barrow, whereas A and C were far to the south, heading southeastward. Lastly, transport for the 26th was a bit more rapid, but with an irregular pattern. Point B was near Alaska after four days, and A, though still south of Kamchatka, was heading rapidly toward Alaska. C and D were over Manchuria and south of Alaska, respectively, each heading southeast.

In summary, then, the results of this trajectory analysis showed that dust originating in the Gobi Desert or the Loess Plateau from 22 through 26 April 1976, could indeed have supplied aerosol to the Barrow atmosphere from the 26th of April through at least the 1st of May, and possibly through the 2nd of May. Such dust was actually observed at Barrow from the 29th of April through at least the 2nd of May. It may have been present earlier, because from the 22nd through the 28th of April no observations could be made because of poor weather at Barrow. There is some question about whether the dust could have been present

over Barrow later than the 2nd, because no observations could be made on the 3rd and on the 4th no haze bands were seen but the measured aerosol levels were high. This point will be treated in more detail below; the main conclusion to be made here is that predicted dust and observed dust at Barrow were in very good agreement.

IV. The dust over Alaska

A. Arctic haze

It was only after the dust cloud reached Alaska that we entered the picture. During April and May 1976 we carried out a field program of direct sampling of the particles constituting bands of Arctic haze, diffuse tropospheric aerosol which occurs north of 70° and at rather high frequencies during clear periods of the year. The presence of such haze in the Arctic atmosphere, which is usually thought to be outstandingly clean, has been a great puzzle since the original notations of its presence by the "Ptarmigan" weather reconnaissance missions over the Arctic Ocean in the 1950's and 1960's. In an attempt to determine the origin of this haze, which we were quite concerned might be anthropogenic, we devised a program to collect haze particles by light aircraft, and to perform chemical, physical, optical, and meteorological analyses, the procedures for and results of which will be presented after a brief introduction to Arctic haze.

One intuitively expects the Arctic atmosphere to have a great purity, because it is so far removed from obvious aerosol sources such as dust storms over continents, wind storms at sea, volcanoes, forest fires, human activities, etc. To a certain extent this is true, as

witnessed by numerous accounts of exceptional visibility. But surface properties of the atmosphere may be deceiving, and can be quite unrepresentative of the atmosphere as a whole. In fact, evidence accumulated over the past 30 years strongly suggests that the Arctic atmosphere is rather turbid, i.e., murky. As mentioned above, first indications of this came from the "Ptarmigan" missions. Mitchell (1956) reported observations of what he called "Arctic haze," an aerosol often found over the Ptarmigan flight paths at altitudes of up to 9000 meters. Vertical visibility was unaffected by Arctic haze; horizontal and slant visibility, however, could be reduced to as little as 3 to 8 km. Mitchell summarized: *"The haze is found during a very high percentage of the time that the weather of the Arctic Ocean is otherwise clear and, being encountered at all normal flight altitudes (700-500-300-mb levels), appears to have considerable vertical depth. Owing to the widely varying conditions of illumination, nothing can confidently be said about the seasonal variation in its frequency. The horizontal extent of the haze is usually limited to segments of the air route which are 500-800 miles (800-1300 km) long, but sometimes stretches over the entire 2000 miles of the reconnaissance track. A curious fact about the haze is that it is seldom found over land, as in the vicinity of Greenland. A ground observer can rarely detect the presence of Arctic haze, just as thin cirrostratus layers encountered by jet aircraft in middle latitudes are often indistinguishable from clear sky to an earth-bound observer. But the similarity between Arctic haze and thin cirrostratus seems to end there: (1) Whereas the cirrostratus appears milky-white from the air, Arctic haze possesses a grey-blue hue in antisolar directions and a*

reddish-brown hue in the direction of the sun. (2) Whereas the cirrostratus is confined to a definite layer in the atmosphere, Arctic haze possesses very diffuse and indistinct upper and lower boundaries and occupies, in the vertical, a considerable fraction of the Arctic troposphere. And finally, (3), whereas the cirrostratus is obviously crystalline when viewed at close range from an aircraft ('You can reach out and touch it'), and produces sun pillars, parhelia, halos and similar optical phenomena in sunlight which are characteristic of ice crystals, the Arctic haze seems completely amorphous to the eye and is almost always without optical effects, at least those which are sufficiently bright to be seen through aircraft windows. The color effects in the haze suggest that, like the low-tropospheric haze of middle latitudes, the constituent particles are no larger than about 2 μ. This size is so small that, should the haze be made up of ice, the cyrstals would be very rudimentary. Since ambient temperatures in the haze are normally in the range of -30 to -35°C, it is yet possible that supercooled water could be present although, to the writer's knowledge, no aircraft icing has ever been reported in Arctic haze."

After this original notation of the existence of Arctic haze, together with a summary of its most obvious visual properties, it seems to have been relegated to the status of a scientific curiosity and forgotten. Then beginning in 1972 a series of radiation measurements conducted by the Geophysical Institute of the University of Alaska revealed unexpectedly high values of turbidity in the Arctic atmosphere and again focused attention on Arctic haze. Furthermore, this research added a new dimension of significance to Arctic haze, that of measurable

radiation effects. Shaw (1975) reported ground-level optical depths near Barrow, Alaska, for 8 and 10 April 1972 of T_o (500 nm) = 0.39 and 0.29 respectively. These values were considered to be abnormally high for this kind of region, inasmuch as they were comparable to or higher than typical mid-latitude values. Similarly high turbidities were also observed for the McCall Glacier in northeastern Alaska during late spring and summer (Shaw and Wendler, 1972). Somewhat lower but highly variable turbidities were found over Barrow in July 1972. For both spring and summer the aerosol concentration decreased exponentially with height, having a scale height of 1.4 $\pm$ 0.3 km.

Unusually high turbidities were again found near Barrow in March 1974 (Holmgren *et al.*, 1974). This time, however, much of the anomalous turbidity was observed to originate with distinct haze layers at altitudes of a few kilometers, a feature of Arctic haze not noted in the original account of Mitchell (1956). Flights in and around these haze layers revealed that they could reduce horizontal visibility to only a few miles. A vertical profile through one such haze layer, obtained by airplane soundings with a sun photometer, is presented as Figure 6 (after Holmgren *et al.*, 1974). The Arctic haze layer is the broad upper layer, roughly one kilometer thick and centered at an altitude of about 1.6 km, below which, at a height of a few hundred meters, is found a more intense but more restricted layer of locally generated ice fog. Figure 6 clearly shows the measurable optical thickness of this Arctic haze layer. This strongly implies that a high frequency of large clouds of Arctic haze ought to have a demonstrable effect on the radiation budget of the Arctic. Indeed, haze layers similar to that of Figure 6 were a

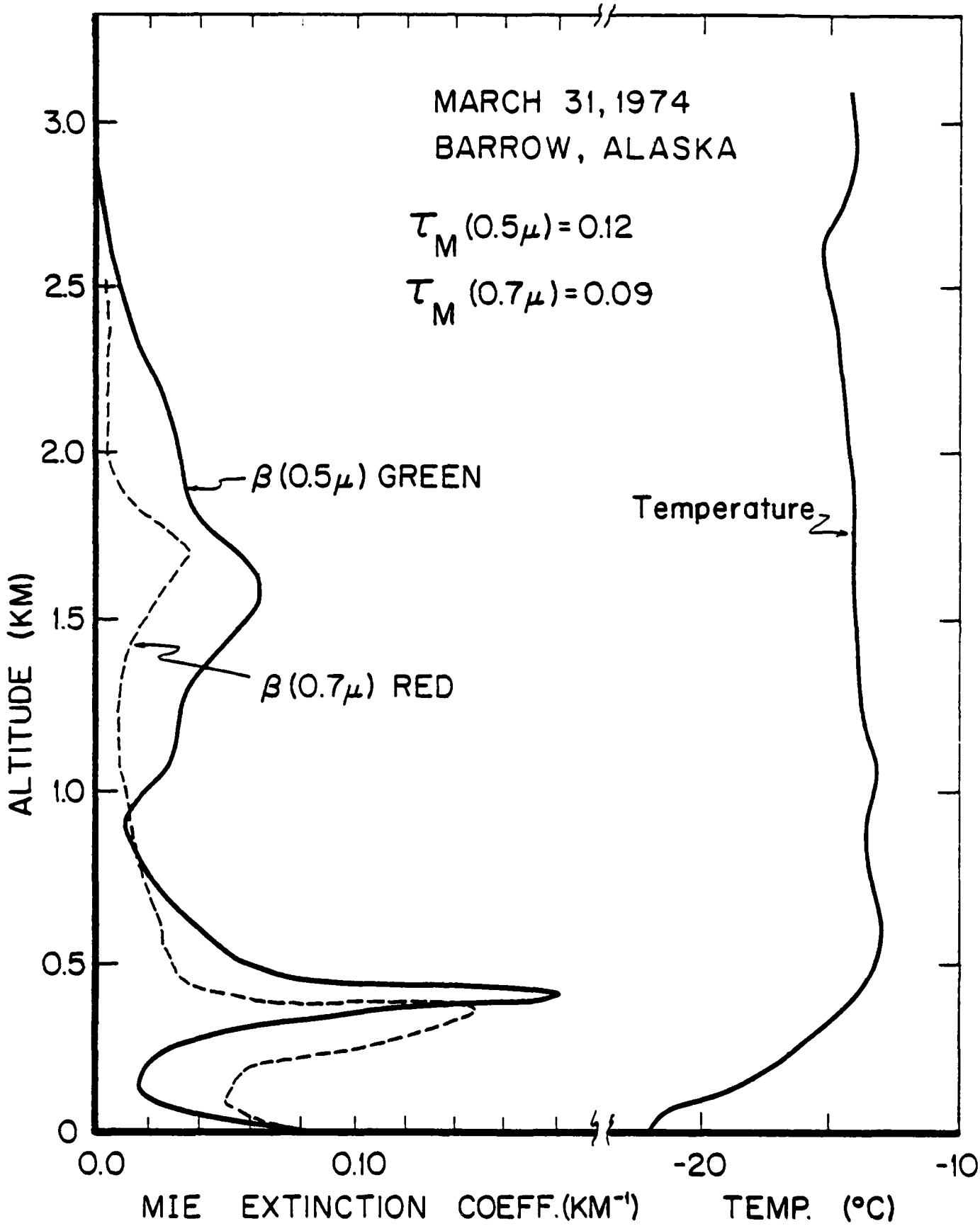


Figure 6. Vertical profiles of aerosol (Mie) extinction coefficients and temperature through an Arctic haze layer (near 2 km). (After Holmgren et al. 1974).

persistent feature of the lower troposphere north of Barrow during the spring of 1974.

Why was the haziness of Alaskan Arctic air unrecognized for so long? After all, optical depths in spring over Barrow are comparable to those found in many medium-to-large cities. Part of the reason is probably because the haze seems to be spread throughout the troposphere, rather than being highly concentrated in the surface layer as it is over cities. Thus, surface visibilities can be very good in the Arctic at the same time that vertical extinction is very strong, i.e., the atmosphere as a whole is very dirty. Vertical extinctions are not obvious to observers without instruments - thus the haze is missed.

The source of Arctic haze is clearly a topic of great potential importance for Arctic climatology as well as for an understanding of the Arctic aerosol itself. We posed several possible sources, both natural and anthropogenic. The list of possible natural sources included (1) local ice-crystal formation from open leads, blowing snow, etc., (2) in situ particle formation from ambient gases, (3) sea salt, (4) continental fragments, and (5) volcanic ash.

Ice crystals (1) were all but ruled out by Mitchell (1956). In situ reaction of ambient ammonia and sulfur dioxide to form stable aerosols (2), such as has been discussed for the stratosphere by Scott and Duffy (1969), and been discounted for the Alaskan lower troposphere by Shaw (1975), who used equilibrium vapor pressures of the products (Lamb, 1971) to suggest that they cannot be formed above a temperature of -60°C. This conclusion is not definitive, however, because winter air temperatures over Alaska do occasionally reach -60°C. Sea-salt

particles (3) can be produced along the coastline near Barrow during the summer half-year when there is open water, but during the spring when Arctic haze is observed the sea is frozen over, hence no sea-salt aerosol can be generated locally. Even when it is produced, it is confined to the marine inversion layer of 1 to 2 km height, whereas Arctic haze is found at all heights in the troposphere. We also considered continental fragments (4), but tended to discount them because they also cannot be produced anywhere near Barrow during the winter when the ground is frozen and snow-covered. Even in the summer the tundra is not a prolific source of dust. Because of the extreme stability of the Arctic atmosphere during winter, it can safely be assumed that Arctic-haze particles above the boundary marine layer must either have been produced *in situ* or have been transported in from great distances.

Very little is known about the contribution of volcanic dust (5) to the Arctic aerosol. The most active volcanoes that inject material into the stratosphere are found in two latitudinal belts, a near-polar belt ($56^{\circ}N$-$65^{\circ}N$) and an equatorial belt ($8^{\circ}S$-$15^{\circ}N$) (Cronin, 1971). There is some evidence of a poleward drift of stratospheric volcanic dust, the particles in which are micron-sized (Lamb, 1968). Stratospheric volcanic dust may last up to ten years in high latitudes (Lamb, 1968). In this regard, it is interesting to note that surface ozone, derived from the stratosphere via the same mechanism as would be expected for surface volcanic dust, shows a pronounced spring maximum (Junge, 1963), in line with the turbidity and haze-layer observations of Shaw (1975) and Holmgren *et al.* (1974).

The University of Alaska observations of Arctic haze raised the question of its possible anthropogenic origin. Shaw (1975) found that high turbidities in April 1972 were associated with air from the south, and low turbidities with air from the north. Trajectory analysis for the persistent haze period of late March and early April 1974 suggested that this air had passed over the highly urbanized east coast of the United States several days earlier. If Arctic haze were really air-pollution aerosol, the implications for man's impact on the Arctic would be great indeed.

B. Sampling and analytical techniques

1. Sampling of haze aerosol aloft

Direct sampling of Arctic-haze aerosol took place during April and early May 1976. The sampling program was based at the Naval Arctic Research Laboratory (NARL) of Barrow, Alaska, and used one of NARL's Cessna 180 single-engine aircraft. A spare right-hand door was obtained and specially modified to hold the sampling probes, as follows: the window was removed and replaced by a two-part assembly, the front half of which was a clear plastic portion of an old DC-3 window, and the rear half of which was an assembly of two PVC plates sandwiched over the window frames, which held the plastic window in place and provided exit ports for the three sampling probes. A single large probe was for an acid-washed 11-cm-diameter Whatman No. 41 cellulose high-volume filter; two smaller probes led to 47-mm diameter Nuclepore and Millipore filters. The Whatman filter collected haze particles for chemical analysis, the Nuclepore filter collected particles for scanning electron microscopy,

SMALLER TYPE

and the Millipore filter collected particles for ice-nucleus counts. All filters were mounted inside the aircraft. Each probe was made isokinetic, so that the sampling process would not discriminate against any particular size of haze particles. This was accomplished by adjusting the diameters of the intake orifices in the cone-shaped nozzles at the front of the probes. Sizes of the orifices depended on the linear velocity through the filters, which was different for all three filters. Holes of approximately the right diameters were drilled at the University of Rhode Island, where the apparatus was constructed, then were modified at NARL based on exact measurements of the performance of each system during a test flight there. The air velocity used to adjust the intake diameter was determined from a pitot tube inserted between the two lower air probes. The aircraft airspeed and altitude simulated actual sampling runs during these tests, so that air velocities into the probes came within 15% of the subsequent values during sampling flights.

Inside the aircraft the filters were mounted in holders which could be quickly attached to or removed from the ends of the PVC probe tubes. Flexible tubing connected each filter holder to its pump. Manometers and flow meters monitored flow rates through the filters. A 1000-watt inverter and four lead-acid batteries powered a Hurricane high-volume pump for the cellulose filters. Smaller pumps for the Nuclepore and Millipore filters were powered directly from the aircraft's electrical and vacuum systems. Pump exhaust was fed out the door; this also provided an extra vacuum which increased the flow rate through the Whatman filter, a very important consideration in aircraft sampling where

sample volumes tend to be very small.

The acid-leaching or "washing" of the Whatman filters was an important part of the overall effort to lower detection limits as much as possible in order to cope with the very small aircraft samples. Although Whatman No. 41 filters have some of the lowest trace-element impurity levels of any air filters available (Dams et al., 1972), these levels can be reduced still further and a significant sampling advantage obtained by judicious acid-washing before sampling. Filters for this experiment were soaked overnight in 2N ultra-pure HNO_3, then rinsed several times with distilled demineralized water until the acidity of the rinse water approached neutrality and its pH remained constant, according to the procedure of Wallace et al.(1976). Final impurity levels of these filters are given in Table 1, and contrasted with the levels in another batch which was not cleaned. Most of the elemental concentrations in the filters were reduced by factors of 2 to 8, but a few elements, e.g. Cl, Cr, Co, Sb, Cs, and Hg remained unchanged in concentration. Much of the success of this experiment must be credited to this washing step.

Contamination of the aircraft samples was considered to be a very serious potential problem, and strict measures were taken to avoid it. There were two possible sources of contamination: engine exhaust being entrained into the probes outside, and cabin air reaching the filters inside. To eliminate leakage from the engine to the outside, all cowling and forward cabin joints were sealed with tape. Tests of nucleus counts at various distances along the wind strut confirmed that no engine exhaust reached the filters. Inside contamination was minimized by keeping

TABLE 1. IMPURITY CONCENTRATIONS IN WASHED AND UNWASHED WHATMAN NO. 41 FILTER PAPER

ELEMENT	WASHED W 41 ng cm^{-2}	UNWASHED W 41 ng cm^{-2}
Na	160 ± 15	270 ± 30
Mg	< 40	< 90
Al	7.3 ± 1.8	30 ± 1
Cl	350 ± 15	370 ± 35
K	< 15	< 120
Sc	0.0010 ± 0.0005	0.0047 ± 0.0006
Ti	< 3	< 12
V	0.026 ± 0.008	0.075 ± .025
Cr	4.9 ± 0.5	4.1 ± 0.1
Mn	0.24 ± 0.02	0.68 ± 0.05
Fe	13 ± 5	33 ± 5
Co	0.040 ± 0.004	0.049 ± 0.005
Ni	< 15	< 4.2
Cu	3 ± 3	< 5
Zn	1.3 ± 0.7	2.5 ± 0.3
Ga	< 0.2	< 0.8
As	< 0.1	< 0.2
Se	< 0.25	< 0.15
Br	2.1 ± 0.1	16.2 ± 1.5
Rb	< 2.5	0.42 ± 0.21
Ag	0.33 ± 0.33	0.041 ± 0.018
Cd	< 1.5	< 2
In	< 0.003	< 0.008
Sb	0.055 ± 0.040	0.038 ± 0.014
I	0.12 ± 0.06	0.58 ± 0.30
Cs	0.10 ± 0.01	0.074 ± 0.009
Ba	< 2.5	< 8
La	< 0.025	< 0.03
Ce	< 0.2	0.10 ± 0.04
Sm	< 0.004	0.006 ± 0.0016
Eu	< 0.01	< 0.01
Lu	< 0.025	< 0.005
Hf	< 0.03	< 0.02
W	0.062 ± 0.018	< 0.5
Au	0.05 ± 0.05	0.019 ± 0.004
Hg	10 ± 4	3.6 ± 0.1
Th	< 0.05	0.015 ± 0.006

the filters off the system and sealed until the sampling altitude had been determined for each flight and sampling was ready to begin; the filters were then quickly opened and immediately attached to the probes. At the conclusion of sampling the process was reversed. Loading of Whatman and Nuclepore filters into and out of their holders for sampling was always done on the ground in a laboratory. Because of small-volume limitation for the Millipore filters, they had to be changed in flight.

There was no evidence of contamination in any of the samples. Measured elemental concentrations, especially on the early flights, were extremely low (Table 4) - some of the lowest that have ever been measured in the Northern Hemisphere. We interpret this as a general absence of contamination. Bromine and chlorine, emitted in engine exhaust as products of combustion of leaded fuel, showed consistently low, near-background concentrations for all flights. In particular, bromine in the external air samples was well over two orders of magnitude lower in concentration than it was in the cabin air (Table 4), which presumably contained small amounts of engine exhaust. The element copper provides a specific and highly sensitive test for contamination of the filters by cabin air. A sample of cabin air showed it heavily loaded with Cu, undoubtedly emitted from the high-volume pump. None of the filter samples showed measurable Cu - thus there was no contamination from cabin air.

Condensation-nuclei counts, which were used to determine the height of the haze layer for each day's sampling, were taken by drawing air in through a small plastic tube which extended out through the window and terminated between the intake nozzles of the Nuclepore and Millipore filters. A hand-operated Gardner counter was used for this purpose.

Radiation measurements were taken with a hand-held sun photometer. This device was pointed directly at the sun's disk through a two-inch hole cut into the clear plastic part of the window of the modified Cessna door. When the sun photometer was not in use the hole was taped over so that no cabin air could escape outside.

Aircraft sampling of Arctic haze aerosol was carried out in April and May of 1976 at NARL, Barrow, Alaska. This period of the year was chosen because it offers the highest probability of clear weather at Barrow; this was also the period when haze layers had been seen in 1972 and 1974. From 12 April to 10 May 1976, one test flight and fourteen sampling flights were made, for a total of 65 hours in the air. Collection altitudes of the haze samples ranged from 1.2 to 3.3 km with an average altitude of about 2 km. We collected nine high-volume cellulose samples, with volumes of 90 to 310 m^3, nine SEM samples with volumes of 6 to 20 m^3, and 55 ice-nucleus samples with volumes of 1 m^3. The individual flights and some comments about each are listed in Table 2. On four of these flights optical measurements were also made; these flights are indicated in Table 2. Concern about very low aerosol concentrations and relatively short sampling flights prompted us to occasionally sample two consecutive flights on a single Whatman high-volume filter and a single Nuclepore filter. This was only done when meteorological conditions indicated that there was no great change in the air mass over Barrow. Therefore, some samples are labelled with two consecutive flight letters, e.g., Sample AB.

The basic philosophy of the aircraft sampling program was to seek to sample haze layers whenever possible. For a first experiment we felt

TABLE 2. DATA FOR FLIGHTS AND AIRCRAFT SAMPLES

FLIGHT	DATE (1976)	FLIGHT TIMES	SAMPLE TIMES	SAMPLE ALTITUDE km	HI VOLUME SCM	PHOTOMETER	NUCLEPORE VOLUME, SCM	REMARKS
A	12-13 April GMT 12 April AST	2317-0344 GMT 1317-1744 AST	0002-0325 GMT 1402-1725 AST	3.4	95.3	No	4.8	Low clouds, fog, light snow at surface. No visible haze. Clear conditions.
B	13-14 April GMT 13 April AST	2005-0035 GMT 1005-1435 AST	2058-0004 GMT 1058-1404 AST	3.4	94.2	No	5.7	Fog at surface, 1200m overcast, 5500m, thin cirrus overcast. Good visibility at sample altitude. No visible haze.
C	14-15 April GMT 14 April AST	1930-0025 GMT 0930-1425 AST	2015-0010 GMT 1015-1410 AST	2.0	151	No	9.2	Atmosphere appeared layered, however, good visibility with scattered clouds at 900m.
D	15-16 April GMT 15 April AST	1945-0030 GMT 0945-1430 AST	2030-0020 GMT 1030-1420 AST	1.2	168	No	9.7	CN counter failed. Used visual observations to choose altitiude. Definite banding seen in atmosphere, chose most defined layer for sampling.
E	17-18 April GMT 17 April AST	2058-0110 GMT 1058-1510 AST	2155-0058 GMT 1155-1458 AST	2.1	130	Yes	7.8	First flight with photometer measurements. Correlation between photometer and CN measurements appears good. A dark band was visible at sample altitude.

TABLE 2. (CONTINUED)

FLIGHT	DATE	FLIGHT TIMES	SAMPLE TIMES	SAMPLE ALTITUDE km	HI VOLUME SCM	PHOTOMETER	NUCLEPORE VOLUME, SCM	REMARKS
F	19-20 April GMT 19 April AST	2035-0100 GMT 1035-1500 AST	2130-0050 GMT 1130-1450 AST	2.1	152	Yes	8.4	No distinct visible banding. C and photometer gave no indication of significant changes in aerosol concentration with height.
G	21 April GMT 21 April AST	1955-2335 GMT 0955-1335 AST	2050-2305 GMT 1050-1305 AST	3.0	86.1	No	5.4	Atmosphere warming aloft. Generally southerly flow. Banding visible.
H	22-23 April GMT 22 April AST	2330-0030 GMT 1330-1430 AST	-- --	--	--	No	--	Flight was terminated due to deteriorating weather one hour after take-off.
I	28,29 April GMT 28 April ADT	2315-0320 GMT 1415-1820 ADT	0002-0305 GMT 1502-1805 ADT	1.8	142	No	7.6	High CN concentrations and deep layer of elevated CN concentration. First time a color has appeared in haze band (light yellow).
J	29 April GMT 29 April ADT	1950-2350 GMT 1050-1450 ADT	2038-2335 GMT 1138-1435 ADT	2.3	120	No	7.4	High CN concentrations and overall hazy conditions. No definite dark banding. Some layering apparent.
K	30 April-1 May GMT 30 April ADT	2005-0100 GMT 1105-1600 ADT	2100-0245 GMT 1200-1545 ADT	2.1	167	No	8.7	A band was visible but not as distinct as J flight. CN concentrations showed a distinct layer.

TABLE 2. (CONTINUED)

FLIGHT	DATE	FLIGHT TIME	SAMPLE TIMES	SAMPLE ALTITUDE km	HI VOLUME SCM	PHOTOMETER	NUCLEPORE VOLUME, SCM	REMARKS
L	1 May GMT	1930-2200 GMT 2315-0120 GMT	2025-2150 GMT 2330-0115 GMT	2.0	138	No	7.5	Very definite visible haze layer, reddish in color, generally seen from Brooks Range to Barrow. Most distinct banding seen.
	1 May ADT	1030-1300 ADT 1415-1620 ADT	1125-1250 ADT 1430-1615 ADT					
M	3-4 May GMT	2010-2330 GMT 0020-0105 GMT	2105-2320 GMT --	2.5-2.8	67.3	Yes	4.9	A well-mixed atmosphere. No distinct banding. Photometer showed no measurable layering. CN concentrations indicated slight increase at 2700m.
	3 May ADT	1110-1430 ADT 1520-1605 ADT	1205-1420 ADT --					
N	5-6 May GMT 5 May ADT	2055-0030 GMT 1155-1530 ADT	2155-0015 GMT 1255-1515 ADT	1.8	91.2	Yes	6.1	Sample represents a deep layer 1800m thick. Most sampling representative of 1800m. Multi-wavelength photometer data was collected. No visible haze, very clear conditions, low CN concentrations.

that these layers offered the best chance of success because (1) they were probably the same material that was more diffusely spread over the rest of the vertical haze area, and (2) the higher concentrations within the layer would give more massive samples than would air outside the layer, which would improve the quality of the analysis. Thus on all flights haze bands were sought according to the following procedure: A particle count was made every 30 seconds or so as the aircraft climbed from the ground to about 3 km, its nominal ceiling. Visual observations were made also. From the particle-count record it was deduced whether there was a haze layer that day and what its altitude was. If there was a layer and the particle counts agreed with visual observations as to its height, the sample was taken at this altitude for as long as the aircraft could remain aloft, usually about 3-1/2 to 4 hours more. If the flight had radiation measurements too, these were compared with the particle counts before the collection altitude was chosen. In general, the agreement between the two types of data was very good. When there was no detectable haze layer, the sample was taken at the altitude of greatest particle concentration.

2. Sampling of surface aerosol

For comparison purposes a series of ground-level samples of the ambient aerosol was taken at the National Oceanic and Atmospheric Administration's Geophysical Monitoring for Climatic Change (GMCC) clean-air sampling site at Barrow. This site lies just to the northeast of NARL, on the east side of Point Barrow and only a few hundred meters from the coastline. A Hurricane high-volume pump, identical to that

used for Whatman sampling in the aircraft, was placed on a small sampling platform upwind (east) of the NOAA observatory building and about 3 m above the snow-covered tundra. Samples were taken on acid-washed Whatman No. 41 filters, for periods ranging from 3 hr to 24 hr. Two of these filters were selected as the most representative of clean-air conditions (with the air coming from a sector from about north through east to nearly south) at Barrow, and were analyzed in the same fashion as the aircraft samples. Sampling data for these filters is given in Table 3.

3. Chemical analysis

The filters were analyzed by nondestructive neutron activation at the University of Rhode Island, in a procedure consisting of two irradiations and five counts. Irradiations were performed in the 2-MW nuclear reactor of the Rhode Island Nuclear Science Center, located on the same grounds as the Graduate School of Oceanography at the University of Rhode Island. The counting chain consisted of an Ortec 38 cm^3 Ge(Li) gamma-ray detector (7% efficiency relative to Na(Tl)), coupled to an Ortec Model 440A amplifier and a Nuclear Data Model 2200 4096-channel analyzer. Spectra were written on magnetic tape by an Ampex Model 7211 7-track recorder and then processed by computer.

Each filter sample was first individually irradiated for five minutes at a thermal-neutron flux of 4×10^{12} n $cm^{-2} sec^{-1}$, cooled for two minutes, and counted for 400 seconds clock-time. After 15 minutes of cooling it was again counted, for 1000 seconds clock-time. From the first count ^{27}Mg, ^{28}Al, ^{51}Ti, ^{52}V, and ^{66}Cu were determined; from the

TABLE 3. DATA FOR THE GROUND-LEVEL SAMPLES

DATE	SAMPLE DURATION	VOLUME, m^3	REMARKS
4/20/76	3 hours	150	Air exclusively from the clean-air sector. Taken between flights F and G.
5/4-5/5/76	24 hours	1200	Air exclusively from the clean-air sector. Taken between flights M and N.

second count ^{24}Na, ^{38}Cl, ^{56}Mn, ^{80}Br, ^{116m}In, ^{128}I, and ^{139}Ba were determined. A pulser was used to correct for dead-time losses during counting. In between the two counts an aluminum flux monitor was counted, which had been co-irradiated with the sample. This served to correct for small variations in the neutron flux from irradiation to irradiation. Standards, consisting of mixtures of known amounts of the elements spotted onto filter paper, were run according to the same sequence and with the same flux monitor.

The filters were then irradiated for 14 hours at the same neutron flux. They were individually wrapped in aluminum foil and all placed into a single polyethylene rabbit together with a standard mixture of the appropriate elements with long-lived nuclides and several aluminum-foil flux monitors along the length of the rabbit. The specific activity of ^{72}Ga was used as the indicator of the relative flux along the rabbit, which in this case varied by about 35% from one end to the other. After a decay of one day the standard and samples were counted for 4000 sec clock-time. From this count ^{24}Na, ^{42}K, ^{69m}Zn, ^{72}Ga, ^{76}As, ^{82}Br, ^{115m}In (from Cd), ^{122}Sb, ^{140}La, ^{153}Sm, ^{152m}Eu, ^{187}W, and ^{198}Au were determined. The samples were counted again after one week for 4000 or 8000 sec, from which count ^{115m}In(from Cd), ^{122}Sb, ^{140}La, ^{153}Sm, and ^{198}Au were again determined. The last count on the samples was performed after approximately one month decay, using counting times of roughly five to 15 hours. From this count ^{46}Sc, ^{51}Cr, ^{59}Fe, ^{60}Co, ^{58}Co(from Ni), ^{65}Zn, ^{75}Se, ^{86}Rb, ^{110m}Ag, ^{124}Sb, ^{134}Cs, ^{131}Ba, ^{141}Ce, ^{152}Eu, ^{177}Lu, ^{181}Hf, ^{203}Hg, and ^{233}Pa(from Th) were determined.

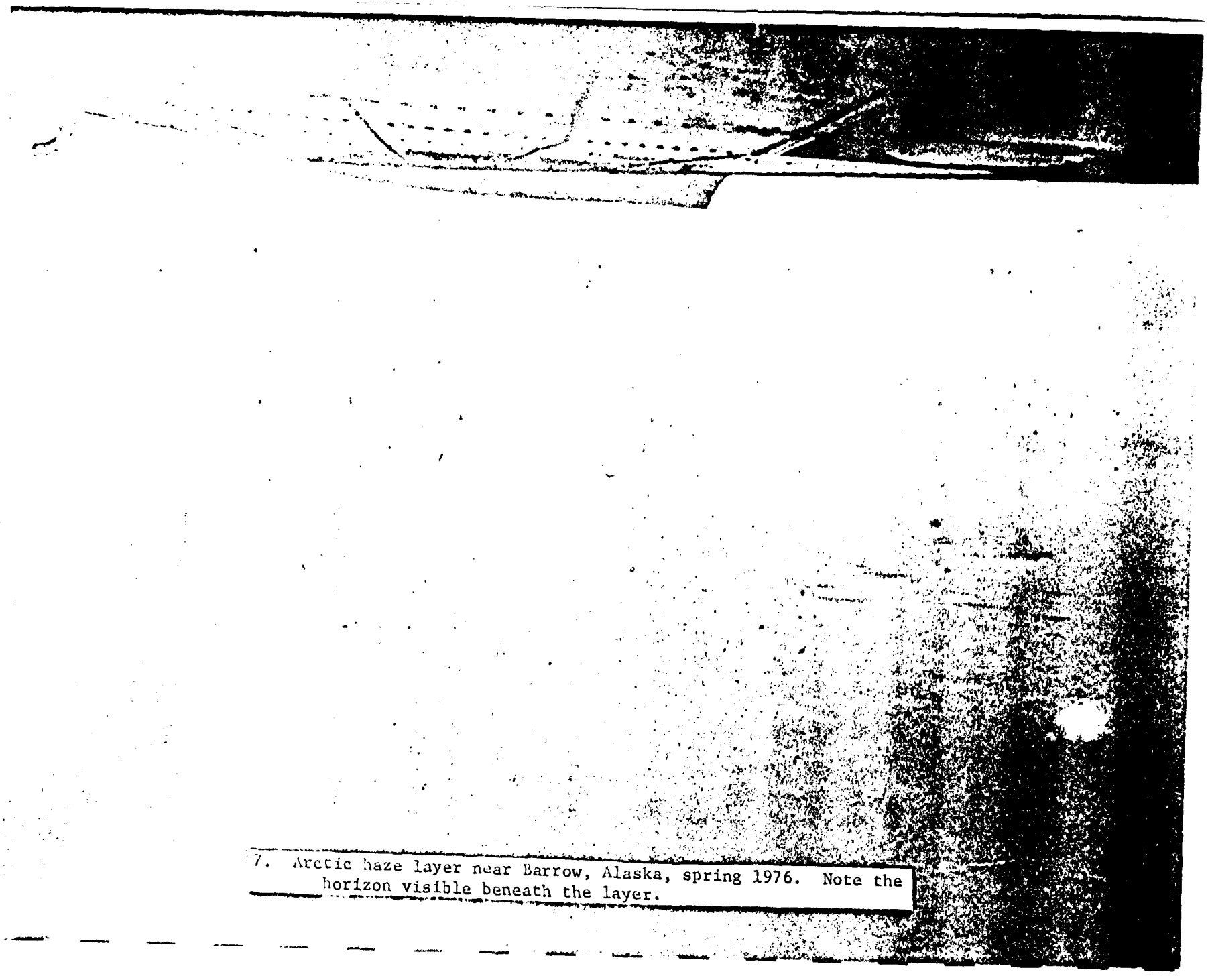
C. Results

1. General

During the course of aircraft sampling a single long-term haze event occurred over Barrow. The visual observations reported in Table 2 are so similar to those from previous years that there was little doubt that the layers seen were the same phenomenon reported by Holmgren et al. (1974). The layers occurred at the same altitudes as before and had the same overall appearance as in previous years. They could be easily seen and photographed. For example, Figure 7 shows a haze layer during flight L, 1 May 1976. The haze band is clearly discernable against the clear-sky background by its darker color and diffuse outlines. Note that the horizon is visible beneath the band.

Although the clarity and intensity of the layers varied from day to day, the continuity of the long-period event was obvious. In fact, this was one of the important findings of this experiment, namely that layers of Arctic haze do not come and go on a day-to-day basis; rather, once they have penetrated the Alaskan Arctic Basin they remain there for several days or more, even while the wind shifts direction. This persistence follows from their large horizontal dimension, commonly reported to be hundreds to thousands of kilometers, which in turn follows from their large-scale origin in Asia and their horizontal mixing and diffusion over the long transport path to Alaska.

The particular haze event observed here began sometime between 23 and 29 April 1976. Before this time some layering of the atmosphere was detected, but condensation-nuclei counts remained low. Between 23 and 29 April, the weather was such that no flights could be made. As of



7. Arctic haze layer near Barrow, Alaska, spring 1976. Note the horizon visible beneath the layer.

flight I, 29 April 1976, high condensation-nuclei counts were observed, together with much more intense general haziness, banding, and coloration of the bands. These optical and particle effects lasted until 3 or 4 May 1976, after which the general haziness and visible bands disappeared and nuclei counts returned to normal. The next flight, M, on 4 May, showed high elemental concentrations (high haze-aerosol mass) but no optical evidence of haze. By flight N, 6 May, elemental concentrations were very low again and the event was over. Depending on whether flight M is considered part of the haze event, its length was therefore somewhere between four to 14 days.

Typical condensation-nuclei profiles for four consecutive flights, the first three of which showed well-developed haze layers, are shown in Figure 8, together with profiles of temperature, dewpoint, and wind velocity. The latter three parameters were obtained from soundings taken at the U.S. Weather Service office in Barrow at 1400 Alaskan standard time (0000 GMT). These observations always fell within the period of each sample flight. Maximum particle concentrations observed were 500 to 600 per cc, which, although not high in absolute terms, are well above the background of 100 to 200 per cc over Barrow.

In flight J, 29 April 1976, a well-definined haze layer was seen between 2 and 2.6 km elevation. There was also some evidence of another, possibly stronger layer above 3 km, where the highest particle counts were observed. Unfortunately, it was not possible to follow this layer any higher because it was above the operational ceiling of the aircraft. Winds aloft were southwesterly. The following day, flight K revealed that the haze layer was somewhat lower and broader, but with

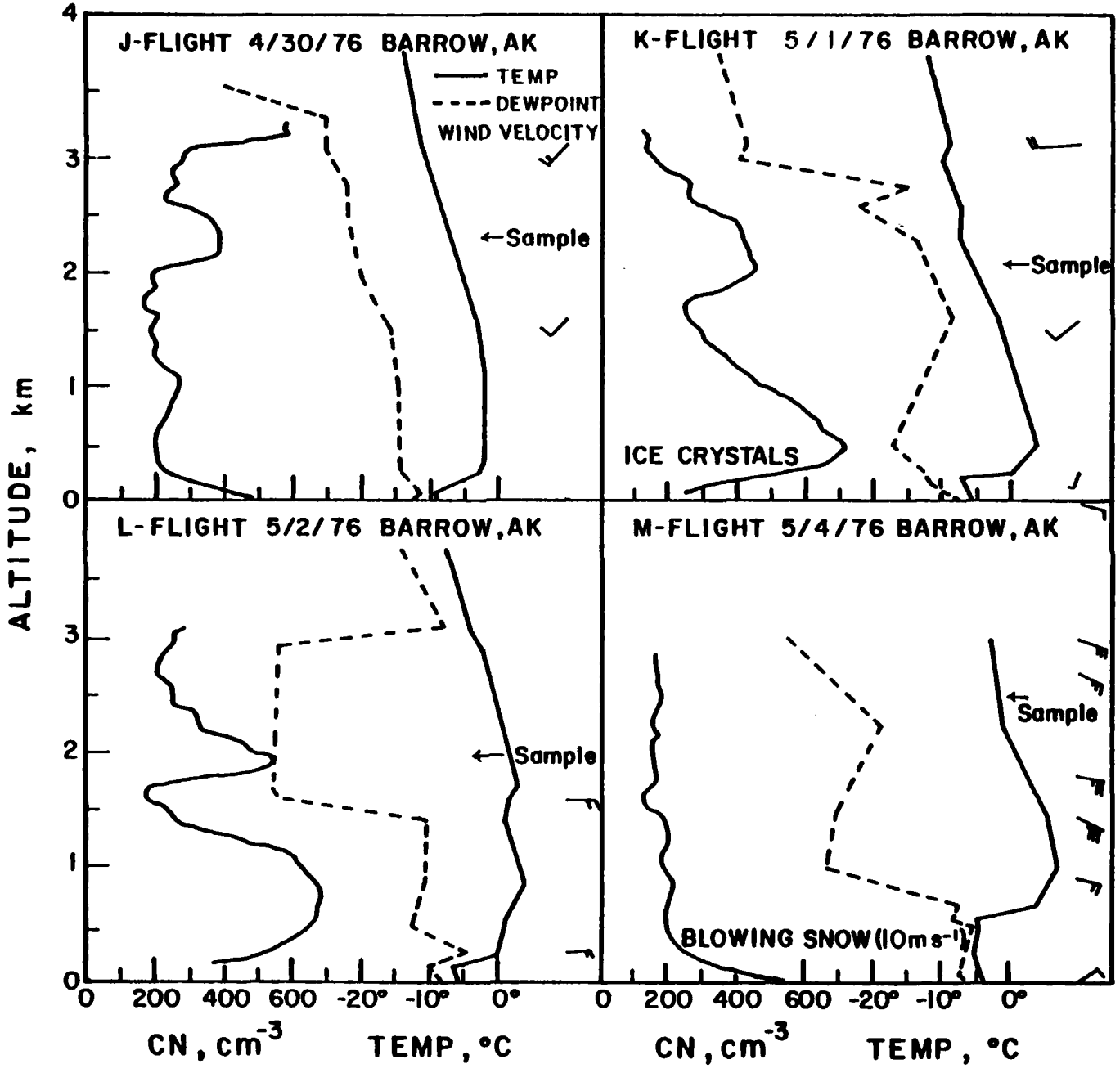


Figure 8. Vertical profiles of particle count, temperature, dewpoint, and wind velocity for selected flights.

approximately the same nucleus count of nearly 400 particles per cc. Note that the lower boundary of a layer of dry air aloft, which could be seen above 3 km in flight J, was also descending. Winds aloft were westerly to southwesterly. By flight L the next day, both the entire dry layer and the haze layer had descended still further and intensified. The haze layer had a much sharper peak than before, between 500 and 600 particles per cc. Winds aloft were then easterly. By flight M two days later, the haze layer had completely disappeared, and the dry air mass was still lower. Winds aloft were easterly to southeasterly. Flight M was the only one of this series that was accompanied by radiation measurements, and these confirmed the absence of a haze layer.

It is interesting that the haze bands were definitely found over the continent, i.e., over Alaska itself. This finding is something of a departure from the earlier reports (Mitchell, 1956) that Arctic haze was found only over the ice. On one occasion during our experiment (Flight L) the haze bands were traced by aircraft all the way south to the Brooks Range. Of course, having originated in Asia and coming to Alaska from the south as the trajectories and local winds aloft have indicated, the Asian dust would have to pass over both the Brooks and Alaskan Ranges on its way to Barrow. In this connection it remains intriguing that Mitchell's account implies that Arctic haze was not found over Alaska itself. Had it really been there, it surely would have been noticed, because the Ptarmigan flights originated in Fairbanks, over which Asian dust is apparently common during the winter. One of us (R. Borys) has seen and photographed bands of Asian dust on a routine commercial flight between Fairbanks and Anchorage during March 1977. The bands were

strikingly visible, to the point that someone with the least idea of what to look for could not possibly have missed them.

Another point of interest in Mitchell's account is that he associated no particular wind direction with the haze bands; in fact, he did not mention winds at all. We, by contrast, found a strong association between Asian dust bands and southerly flow. (As noted above, temporary easterly or westerly winds did not seem to affect the haze bands greatly. Northerly winds, however, never brought in strong haze bands during our measurements, although some banding was seen under these conditions(flights C and D, for example). It may well be possible that at other times of the year haze bands come from the north, from a source other than the Asian deserts.) Could it be that our Asian dust bands are not the same Arctic haze that Mitchell described? The answer is not yet clear. The end of this article will address this problem in more detail.

These southerly winds aloft which carried the Asian dust to Barrow manifested themselves in rising temperatures both aloft (from -27C in flight B to +2C in flight L) and at the surface (during the haze event there was melting snow in Barrow, the first local thaw of the season). This temperature effect is discussed again at the end of the article as evidence for the penetration of southern air over Barrow.

Light winds seemed to favor the maintenance of a haze layer's integrity and high winds seemed to break it up, as, for example, on flight M where elemental concentrations were high, but no layer was detected otherwise. The haze layers also appeared to be associated with dry air masses, the lower boundary of which was found at progressively

lower heights during the course of the event (see Figure 8). This dryness was probably a direct consequence of the dry air masses under whose influence the dust was generated in Asia.

2. Elemental composition

The composition of the haze layers was crustal, i.e., natural rather than anthropogenic. It was this finding that originally prompted us to seek a source for this aerosol in arid regions where crustal dust could be generated in abundance, and ultimately pointed to the Asian Deserts. For this reason the chemical data will now be discussed in some detail.

The results of neutron-activation analysis of the nine high-volume haze-layer filter samples, as well as of the cabin-air and two ground-level samples, are presented in Table 4 as ng/SCM (25C, 1013 mb). It can be seen from this table that 16 to 29 elements were normally determinable in each flight sample, except for N, where only 10 elements could be determined. Haze-layer samples IJ and KL had the largest number of determinable elements in them, because they had the best combination of high concentrations and large air volumes sampled. Scrutiny of this table reveals that the great majority of these elements show qualitatively similar behavior during the haze event; that is, they increase in concentration by roughly an order of magnitude, in some cases as much as a factor of 20 or more, then decrease to nearly their original levels after the event. There are, however, exceptions to and gradations in this behavior. For example, Al and Sc increase by factors of 20 to 30 from sample AB to sample IJ, whereas V and As increase only by

ELEMENT	AB	CD	E	F	G	IJ
Na	17.6 ± 4.1	28 ± 3	12.0 ± 5.8	21 ± 5	50 ± 10	61 ± 5
Mg	12 ± 7	16 ± 7	26 ± 11	< 35	76 ± 40	110 ± 30
Al	13.1 ± 1.3	18.2 ± 1.1	40 ± 2	39 ± 2	116 ± 7	280 ± 10
Cl	42 ± 8	19.7 ± 4.7	< 35	< 35	69 ± 18	8 ± 6
K	8.7 ± 2.1	11.2 ± 1.9	9.3 ± 2.5	21 ± 4	56 ± 8	93 ± 5
Sc	0.00139 ± 0.00007	0.0027 ± 0.0004	0.0073 ± 0.0010	0.0062 ± 0.0007	0.0153 ± 0.0017	0.042 ± 0.004
Ti	3.2 ± 0.5	1.68 ± 0.38	2.3 ± 0.7	3.1 ± 0.7	10.8 ± 1.5	21 ± 1.7
V	0.046 ± 0.008	0.158 ± 0.011	0.144 ± 0.014	0.108 ± 0.012	0.28 ± 0.03	0.47 ± 0.04
Cr	< 7	< 1	< 3	< 12	< 8	< 2
Mn	0.192 ± 0.016	0.38 < 0.02	0.67 ± 0.05	0.67 ± 0.04	1.97 ± 0.10	3.5 ± 0.3
Fe	11.8 ± 3.9	8.1 ± 2.2	29 ± 7	19.6 ± 3.5	82 ± 9	130 ± 10
Co	< 0.04	< 0.01	0.013 ± 0.011	< 0.02	0.027 ± 0.013	0.047 ± 0.006
Ni	< 13	< 3	< 7	< 7	< 8	< 2
Cu	< 4	< 3	< 7	< 7	< 10	< 7
Zn	< 4	< 0.7	< 2	< 1.8	1.57 ± 0.76	1.32 ± 0.30
Ga	< 0.08	< 0.07	< 0.1	< 0.14	< 0.2	< 0.1
As	0.029 ± 0.012	0.090 ± 0.009	0.105 ± 0.016	0.077 ± 0.015	0.20 ± 0.03	0.21 ± 0.02
Se	0.139 ± 0.032	< 0.1	0.085 ± 0.037	< 0.1	< 0.3	0.080 ± 0.017
Br	1.62 ± 0.13	3.0 ± 0.1	2.0 ± 0.2	2.2 ± 0.2	3.9 ± 0.3	2.6 ± 0.2
Rb	< 4	< 0.7	< 1	< 0.7	< 1	0.44 ± 0.18
Ag	< 1.6	< 0.3	< 0.7	< 0.7	< 1	< 0.4
Cd	< 1.3	< 0.5	< 0.7	< 0.7	< 1	0.9 ± 0.2
In	< 0.0013	< 0.002	< 0.003	< 0.003	< 0.008	0.0019 ± 0.0011
Sb	< 0.07	0.031 ± 0.011	< 0.1	0.023 ± 0.014	< 0.1	0.032 ± 0.014
I	0.32 ± 0.04	0.56 ± 0.03	0.54 ± 0.06	0.65 ± 0.05	0.68 ± 0.09	0.71 ± 0.05
Cs	< 0.08	< 0.01	< 0.03	< 0.02	< 0.05	0.029 ± 0.006
Ba	0.54 ± 0.21	0.43 ± 0.20	< 2	< 2	2.0 ± 0.8	2.4 ± 0.2
La	< 0.013	< 0.007	0.0162 ± 0.008	0.0173 ± 0.0058	0.048 ± 0.013	0.147 ± 0.012
Ce	< 0.4	0.020 ± 0.011	< 0.1	0.066 ± 0.020	< 0.1	0.27 ± 0.04
Sn	0.00171 ± 0.00020	0.00123 ± 0.00021	0.0032 ± 0.0006	0.00199 ± 0.00030	0.0083 ± 0.0013	0.0199 ± 0.0005
Eu	< 0.007	< 0.003	< 0.007	< 0.007	< 0.01	0.0066 ± 0.0012
Lu	< 0.03	< 0.003	< 0.007	< 0.007	< 0.008	0.0029 ± 0.0012
Hf	< 0.04	< 0.01	< 0.01	< 0.01	0.014 ± 0.006	0.0103 ± 0.0019
W	0.114 ± 0.025	< 0.05	< 0.1	< 0.1	< 0.1	< 0.1
Au	< 0.07	< 0.03	< 0.1	< 0.07	< 0.1	< 0.06
Hg	< 26	< 3	< 7	< 7	< 13	< 3
Th	< 0.05	< 0.007	< 0.01	0.0119 ± 0.0039	0.027 ± 0.008	0.045 ± 0.004

TABLE 4. Elemental Concentrations, ng/SCM, in Filter Samples

KL	M	N	CABIN AIR	GROUND SAMPLES 3-HOUR	GROUND SAMPLES 24-HOUR
33 ± 4	43 ± 12	< 25	240 ± 30	127 ± 14	179 ± 10
94 ± 7	180 ± 40	< 40	< 450	< 30	41 ± 5
240 ± 10	320 ± 20	17.4 ± 2.0	600 ± 30	37 ± 2	18.1 ± 0.5
5.7 ± 4.8	< 80	< 60	620 ± 70	143 ± 10	166 ± 13
79 ± 5	109 ± 11	7.9 ± 3.2	168 ± 45	25 ± 7	23 ± 4
0.036 ± 0.004	0.051 ± 0.004	0.0023 ± 0.0007	0.048 ± 0.006	0.0031 ± 0.0005	0.0025 ± 0.0003
15.3 ± 1.4	19.1 ± 2.0	2.1 ± 0.6	98 ± 20	< 5	0.92 ± 0.41
0.44 ± 0.02	0.53 ± 0.05	0.023 ± 0.01	1.24 ± 0.17	0.45 ± 0.02	0.24 ± 0.01
< 4	< 4	< 12	33 ± 10	< 6	< 1
3.7 ± 0.2	4.5 ± 0.3	0.25 ± 0.03	8.0 ± 0.4	0.63 ± 0.03	0.52 ± 0.02
112 ± 9	145 ± 20	< 70	370 ± 50	25 ± 4	10.7 ± 1.1
0.043 ± 0.006	0.040 ± 0.020	< 0.1	0.33 ± 0.06	0.097 ± 0.009	0.27 ± 0.02
< 2	< 14	< 25	< 90	5.6 ± 1.3	1.50 ± 0.31
< 6	< 20	< 12	4600 ± 100	29 ± 2	31 ± 1
0.91 ± 0.24	< 3.3	< 7	51 ± 5	11.8 ± 0.9	51 ± 5
< 0.07	< 0.3	< 0.2	< 0.2	< 0.3	< 0.1
0.146 ± 0.011	0.169 ± 0.028	0.02 ± 0.02	< 1	< 0.1	< 0.03
< 0.07	< 0.3	< 0.6	< 3	0.141 ± 0.048	0.038 ± 0.006
2.4 ± 0.1	2.1 ± 0.3	2.2 ± 0.3	630 ± 40	19 ± 1	11.7 ± 0.5
0.30 ± 0.10	< 3	< 5	< 40	< 3	< 0.6
< 0.4	< 2	< 4	< 7	< 3	< 0.4
< 0.2	< 1.5	15 ± 1	44 ± 12	2.6 ± 0.3	4.6 ± 0.7
< 0.002	< 0.015	< 0.004	< 0.07	< 0.003	0.00074 ± 0.00037
0.0111 ± 0.0050	< 0.2	< 0.2	6.8 ± 0.4	0.46 ± 0.03	0.085 ± 0.006
0.66 ± 0.04	1.32 ± 0.12	0.65 ± 0.08	1.44 ± 0.66	0.71 ± 0.06	0.48 ± 0.03
0.0146 ± 0.0045	< 0.08	< 0.1	< 0.6	< 0.06	< 0.02
2.4 ± 0.2	2.5 ± 0.7	< 2	< 60	3.5 ± 0.5	< 2
0.130 ± 0.007	0.165 ± 0.013	< 0.02	0.33 ± 0.06	0.021 ± 0.007	0.0125 ± 0.0031
0.20 ± 0.03	0.21 ± 0.04	< 0.1	0.62 ± 0.17	< 0.3	0.0134 ± 0.0035
0.0164 ± 0.0006	0.024 ± 0.002	< 0.004	< 0.02	< 0.002	0.00163 ± 0.00021
0.0032 ± 0.0012	< 0.03	< 0.01	< 0.14	< 0.03	0.00033 ± 0.00021
0.00141 ± 0.00056	< 0.008	< 0.05	< 0.2	< 0.04	< 0.004
0.0066 ± 0.0017	< 0.033	< 0.07	< 0.5	< 0.06	0.00136 ± 0.00060
< 0.05	< 0.2	< 0.15	< 1	< 0.11	< 0.06
< 0.04	< 0.16	< 0.1	0.80 ± 0.06	< 0.06	< 0.01
< 3	< 16	< 40	26 ± 8	< 30	< 6
0.035 ± 0.004	0.053 ± 0.008	< 0.07	0.080 ± 0.024	< 0.06	0.00147 ± 0.00066

TABLE 4. (CONTINUED)

about a factor of 10, and Cl, Br, I, Se, and Sb essentially show no increase at all. Why should this be?

The answer can be seen by considering the aerosol-crust enrichment factors for the various elements in the samples. The aerosol-crust enrichment factor for an element X in an aerosol is defined by

$$EF_X = (X/Al)_{aerosol}/(X/Al)_{crust}$$

where X/Al refers to the ratio of concentrations of element X and Al in the crust (here taken as average crustal rock as quoted by Mason, 1966) and in aerosol. Aluminum is used as reference element for a number of reasons: it is abundant in the crust (81,300 ppm in average crustal rock, for example), is easily determined by neutron activation even at very low concentrations, is easily determined by other analytical techniques (atomic absorption, for example), has very few specific pollution sources, and is generally free from contamination during sampling. For a fuller discussion of enrichment factors and their significance in aerosol chemistry, see Rahn (1976b).

Elemental enrichments for the samples of this experiment are listed in Table 5. They range from a low of about 0.4 for Na to a high of more than 10,000 for several elements. Such a range is quite typical for aerosols in general. Approximately one-half of the elements have enrichments near to unity, whereas the other elements have much higher values, between about 10 and 10,000 or more. This is also typical of the world aerosol (Rahn, 1976b). It is interesting to note that these two groups of elements behave quite differently over the series of flights. The low-enrichment group, typified by Al but also containing

TABLE 5 . AEROSOL-CRUST ENRICHMENT FACTORS

ELEMENT	FLIGHTS									CABIN	GROUND SAMPLES	
	AB	CD	E	F	G	IJ	KL	M	N	AIR	3-HR	24-HR
Na	3.8	4.5	0.86	1.50	1.23	0.62	0.40	0.40	<4	1.17	9.8	28
Mg	3.5	3.4	2.6	<4	2.6	1.46	1.54	2.3	<8	<3	<3	8.8
Al	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Cl	2000	680	<600	<600	370	18.2	15.0	<160	<2200	650	2400	5700
K	2.1	1.94	0.73	1.68	1.52	1.03	1.04	1.09	1.43	0.88	2.1	3.9
Sc	0.39	0.54	0.68	0.59	0.49	0.55	0.55	0.60	0.48	0.29	0.30	0.50
Ti	4.4	1.71	1.06	1.47	1.73	1.34	1.18	1.12	2.3	3.0	<2.5	0.94
V	2.1	5.2	2.2	1.66	1.46	1.00	1.11	1.00	0.79	1.25	7.2	7.8
Cr	<410	<50	<70	<250	<60	<5	<15	<11	<600	45	<125	<50
Mn	1.25	1.81	1.44	1.46	1.46	1.07	1.33	1.21	1.22	1.14	1.45	2.5
Fe	1.46	0.73	1.16	0.81	1.14	0.75	0.76	0.75	<7	0.99	1.10	0.96
Co	<10	<2	1.02	<2	0.75	0.54	0.59	0.41	<25	1.81	8.5	48
Ni	<1100	<200	<200	<200	<80	<9	<12	<50	<1500	<150	163	89
Cu	<450	<270	<260	<300	<130	<40	<40	<100	<1000	11,200	1150	2500
Zn	<350	<50	<70	<60	15.7	5.4	4.4	<15	<500	99	370	3300
Ga	<35	<20	<16	<20	<12	<2.3	<2	<5	<75	<1.5	<40	<30
As	99	220	119	89	79	34	28	24	61	<84	<120	<80
Se	17,200	<10,000	3400	<5000	<4000	460	<500	<1500	<60,000	<10,000	6100	3400
Br	4000	5400	1650	1820	1110	300	330	220	4100	34,000	16,300	21,000
Rb	<280	<35	<30	<16	<10	1.39	1.13	<8	<250	<50	<70	<35
Ag	<140,000	<21,000	<20,000	<21,000	<15,000	<1450	<2000	<8000	<250,000	<13,000	<90,000	<30,000
Cd	<41,000	<12,000	<7,000	<8,000	<5,000	1360	<500	<2000	360,000	30,000	28,000	103,000
In	<90	<100	<70	<80	<60	5.4	<10	<40	<170	<100	<60	33
Sb	<2100	690	<1200	240	<500	46	27	<3000	<5000	4600	5000	1910
I	3900	5000	2200	2700	950	410	450	680	6100	390	3100	4300
Cs	<170	<20	<24	<16	<12	2.8	1.65	<7	<200	<25	<40	<30
Ba	7.8	4.5	<11	<12	3.4	1.59	1.91	1.51	<30	<20	18.2	<25
La	<3	<2	1.10	1.20	1.13	1.41	1.48	1.41	<4	1.51	1.56	1.87
Ce	40	1.51	<4	2.3	<1.5	1.30	1.13	0.89	<10	1.41	<10	1.00
Sm	1.76	0.92	1.08	0.69	0.97	0.95	0.93	1.04	<3	<0.5	<0.7	1.22
Eu	<35	<12	<12	<12	<8	1.58	0.91	<6	<50	<20	<50	1.22
Lu	<330	<30	<30	<30	<11	1.69	0.96	<5	<500	<70	<170	<40
Hf	<90	<20	<8	<8	3.3	0.98	0.75	<3	<120	<20	< 40	2.0
W	470	<160	<160	<160	<60	<23	<11	<40	<500	<100	<170	<200
Au	<100,000	<40,000	<60,000	<40,000	<25,000	<4300	<4000	11,000	<150,000	27,000	<35,000	<12,000
Hg	<2,000,000	<180,000	<200,000	<200,000	<150,000	<13,000	<13,000	<60,000	<2,100,000	43,000	<800,000	<400,000
Th	<50	<4.0	<4	3.4	2.6	1.79	1.64	1.88	<50	1.51	<17	0.91

Sc, Mn, Fe, the rare earths, and Th, is just the group that increases in concentration by large factors over the flights, particularly at the onset of the strong haze layers in sample IJ. In contrast, the high-enrichment group of elements, typified by As but also containing Sb, Se, Cl, Br, and I, is just the group that shows the least variation in concentration over the flights, and in particular, only a small increase in the strong haze layers. The elements Na, K, Ba, and V seem to be intermediate in behavior, having moderate enrichments and for the most part moderate increases in concentration in the haze layers. Figure 9 shows the concentration trends of these two groups of elements, represented by Al and As, respectively, together with the ranges of their enrichment factors over the nine aircraft samples.

As a consequence of the fact that the enriched elements increased only relatively little in concentration during the flights, their enrichments decreased on the average over the series, especially at the onset of the haze. The nonenriched elements, on the other hand, increased together, so had more nearly constant enrichment factors. Intermediate elements behaved intermediately. Figure 10 shows these trends graphically for the nonenriched elements Al and Sc, the intermediate element Ba, and the enriched element As. It is clearly seen here that Sc had no systematic change of enrichment factor, whereas Ba and As showed moderate and large decreases of enrichment, respectively. In general it seemed that the larger the enrichment factor, the more it decreased in the haze.

A similar depression of enrichment factors in the haze aerosol can be seen relative to average remote continental northern aerosol. To demonstrate this, we have calculated weighted mean concentrations and

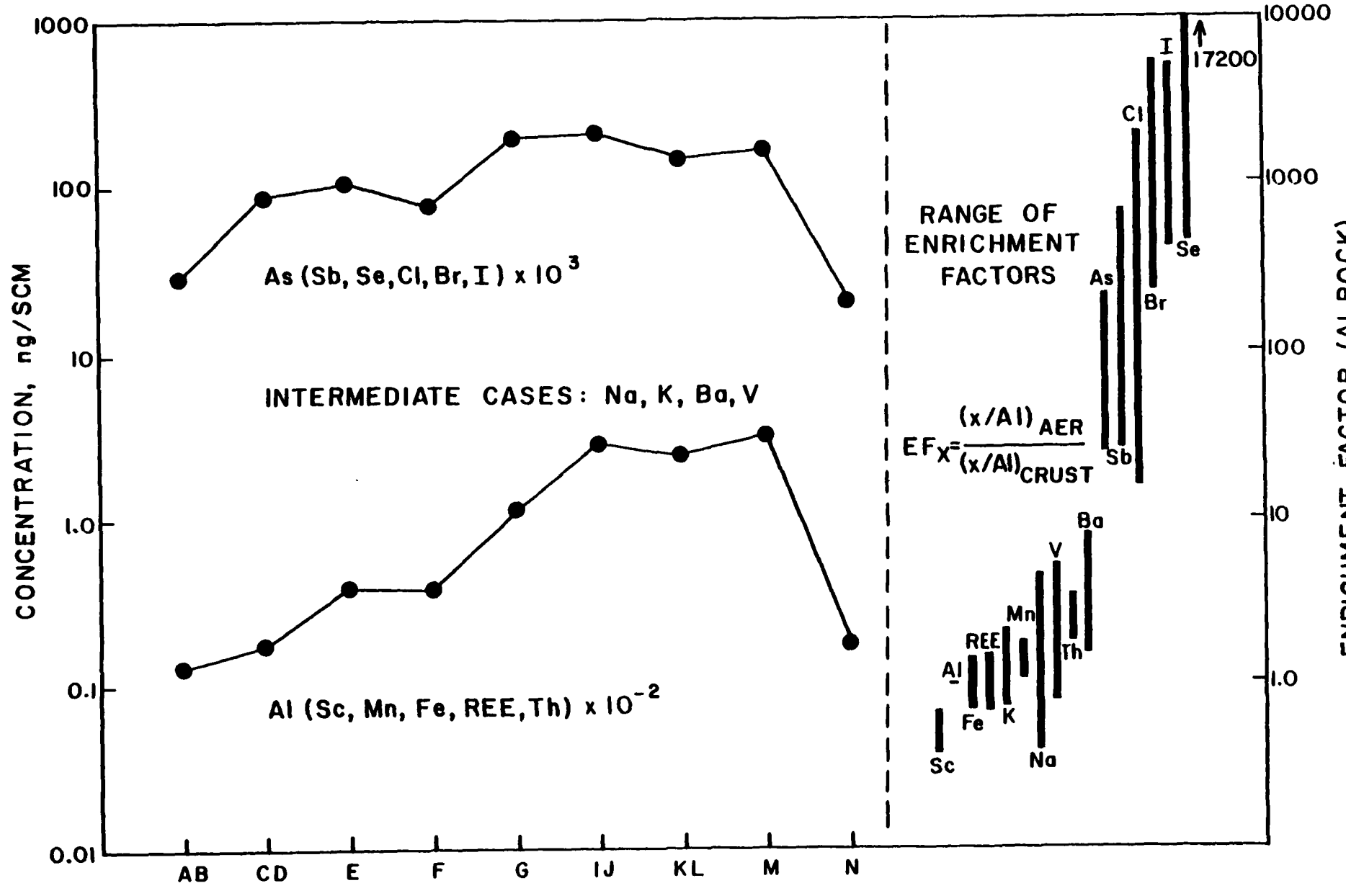


Figure 9. Concentration patterns for crustal and enriched elements over the series of flights at Barrow; ranges of enrichment factors for various elements in these samples.

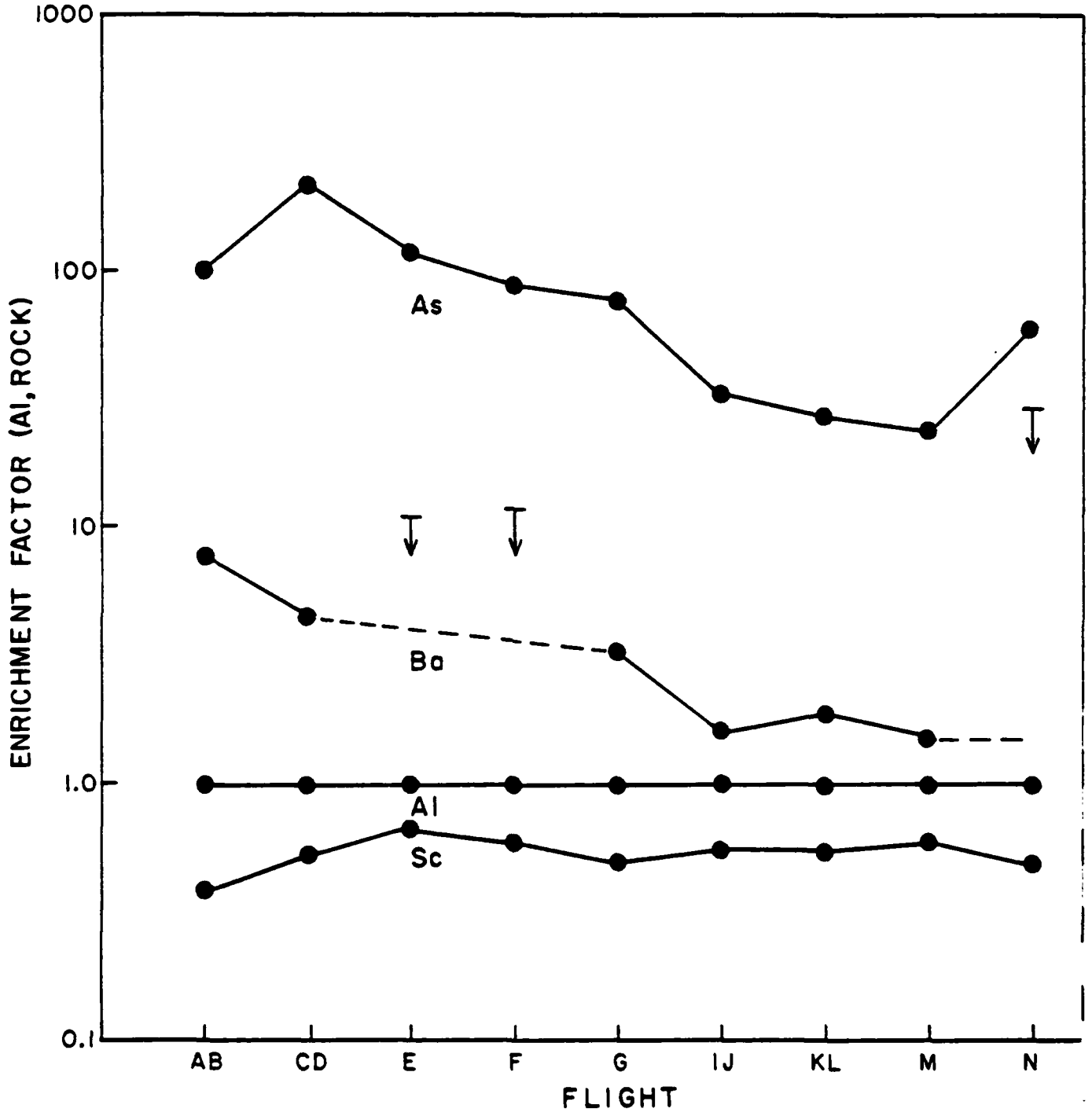


Figure 10. Enrichment-factor trends for Al, Sc, Ba, and As over the sampling period.

enrichment factors for the haze aerosol of samples IJ, KL, and M. Results are given in Table 6. These enrichment factors are compared to those of average remote northern continental aerosol (Rahn, 1976b) in Figure 11. As in Figure 10, nonenriched elements have similar enrichments in haze and nonhaze aerosols, but enriched elements have systematically lower enrichments in Arctic haze. Again, the higher the enrichment of an element in the normal aerosol, the more it is depressed in Arctic haze.

Taken together, the above observations suggest that Arctic haze is much more strongly crustal than is normal northern aerosol. The following argument, based on the behavior of the element vanadium in the samples, demonstrates that Arctic haze is essentially exclusively crustal, i.e., natural rather than pollution-derived. This argument is based on our present knowledge of only two major sources for atmospheric V: the crust and combustion products of residual fuel oil (Zoller et al., 1973). In remote areas free of significant air pollution the V/Al ratio is very close to that of the bulk crust. In other words, the enrichment factor of V is unity. In urban areas, on the other hand, the V enrichment factor is much higher, varying from 5 to 500 depending on the area and its source of residual oil (Rahn, 1976b). The northeast United States, for example, uses Venezuelan residual oils which are unusually rich in V, and hence has abnormally high V enrichments in its aerosol. Beside the crust, no other major natural sources of V are known. Thus, V enrichment factors greater than about 1.5 or so signify the presence of air pollution, and V enrichments below 1.5 mean that the aerosol is mostly crustal.

TABLE. 6 WEIGHTED MEAN ELEMENTAL CONCENTRATIONS
AND ENRICHMENT FACTORS FOR HAZE SAMPLES IJ, KL, and M

ELEMENT	CONCENTRATION*, ng/SCM	ENRICHMENT FACTOR
Na	46 ± 14	0.49
Mg	118 ± 36	1.70
Al	270 ± 30	1.00
Cl	7** ± 4	16
K	91 ± 13	1.06
Sc	0.041 ± 0.006	0.56
Ti	18.3 ± 2.9	1.25
V	0.47 ± 0.04	1.05
Cr	< 4	<12
Mn	3.8 ± 0.4	1.20
Fe	126 ± 14	0.76
Co	0.044 ± 0.005	0.53
Ni	< 2	<8
Cu	< 7	<40
Zn	1.12** ± 0.29	4.8
Ga	< 0.1	<2
As	0.176 ± 0.032	29
Se	< 0.1	<600
Br	2.4 ± 0.2	290
Rb	0.37** ± 0.10	1.24
Ag	< 0.4	<1800
Cd	< 1	<1500
In	< 0.002	<6
Sb	0.024** ± 0.011	36
I	0.81 ± 0.28	490
Cs	0.022** ± 0.010	2.2
Ba	2.4 ± 0.2	1.70
La	0.144 ± 0.015	1.45
Ce	0.23 ± 0.04	1.15
Sm	0.0193 ± 0.0032	0.97
Eu	0.0049** ± 0.0024	1.23
Lu	0.0022** ± 0.0011	1.32
Hf	0.0084** ± 0.0026	0.84
W	< 0.1	<20
Au	< 0.1	<8000
Hg	< 3	<12,000
Th	0.043 ± 0.008	1.80

*Standard deviation is the larger of analytical uncertainty and actual dispersion
**From IJ, KL only

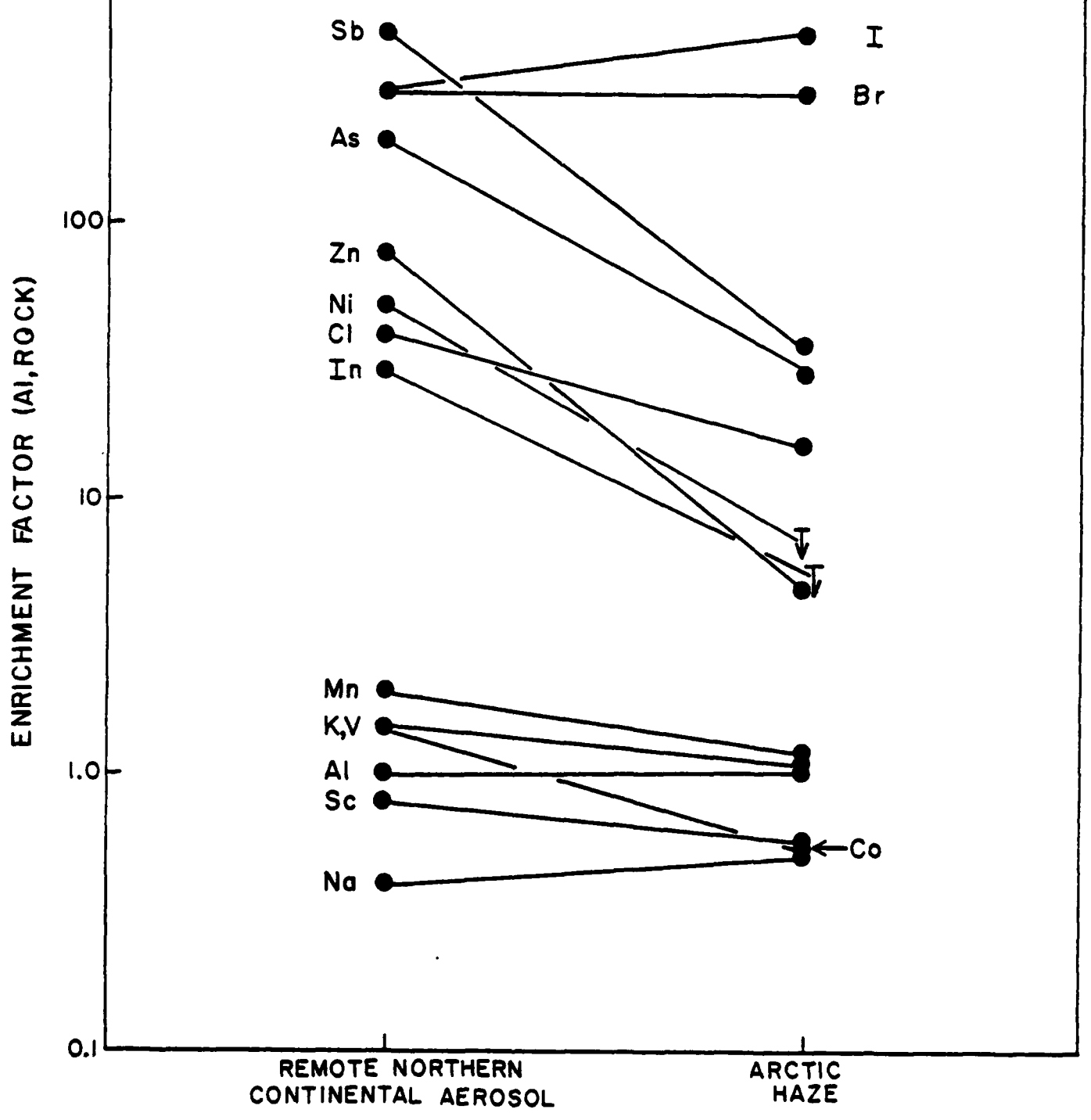


Figure 11. Elemental enrichment factors in Arctic haze of spring 1976 versus remote northern continental aerosol (Rahn, 1976b).

Figure 12 shows the concentrations and enrichments of " and Al in the nine aircraft samples. In the early flights, which represent typically "clean" Arctic air with very low concentrations of both V and Al (and most of the other elements as well), the V enrichment factors ranged between 2 and 5, which definitely indicated that the V was pollution-derived. But with the onset of the haze the enrichment factors for V decreased sharply, and during the peak of the haze they were essentially 1.0. This meant unambiguously that this Arctic haze was crustal (i.e., natural) aerosol rather than pollution aerosol. In particular, it could not possibly have come from the northeast United States or from Europe.

The fact that the composition of the Arctic haze bands is natural is made all the more interesting because it is set against a background of air pollution. As noted above, the V enrichment factors of 2 to 5 in samples A through E indicate air pollution. Because of the altitude of these samples and the lack of emissions of pollution V in the Arctic, this air pollution must also be the result of moderate- to long-range atmosphere transport. This idea is nicely confirmed by backwards trajectory analysis from Barrow for the sampling period, as shown in Figure 13 for the 700-mb level. Only haze-containing air had recently passed over the Asian Deserts; for air arriving at Barrow before and after the haze event the trajectories are quite different. Specifically, prehaze aerosol came from the north, with air of the 15th through the 19th April coming particularly rapidly from over the pole. The times of these air masses correspond to samples CD and E, which had the highest V enrichment factors of the series. These air masses had probably originated

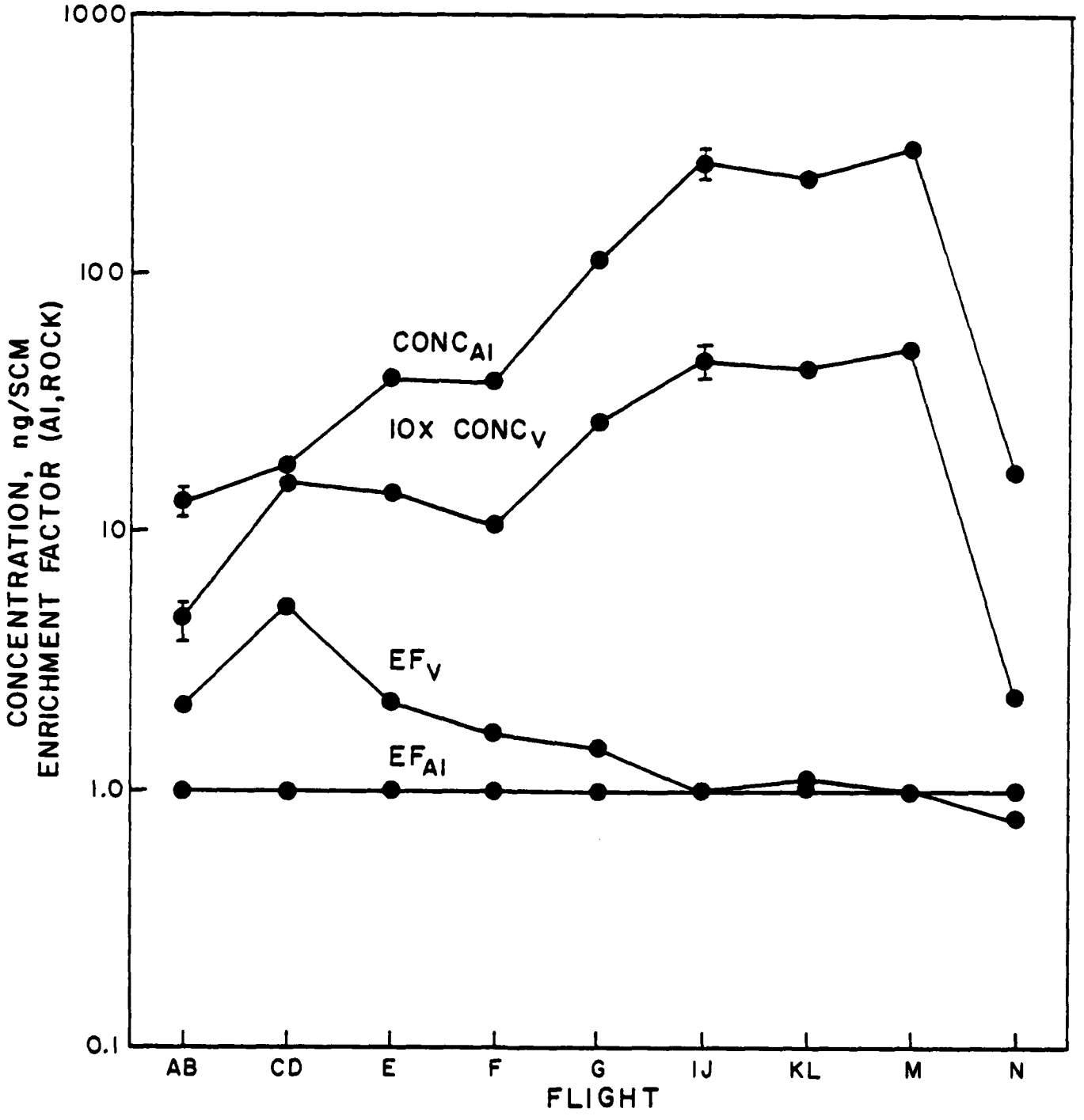


Figure 12. Concentration and enrichment factors of V and Al in aerosol over Barrow, spring 1976.

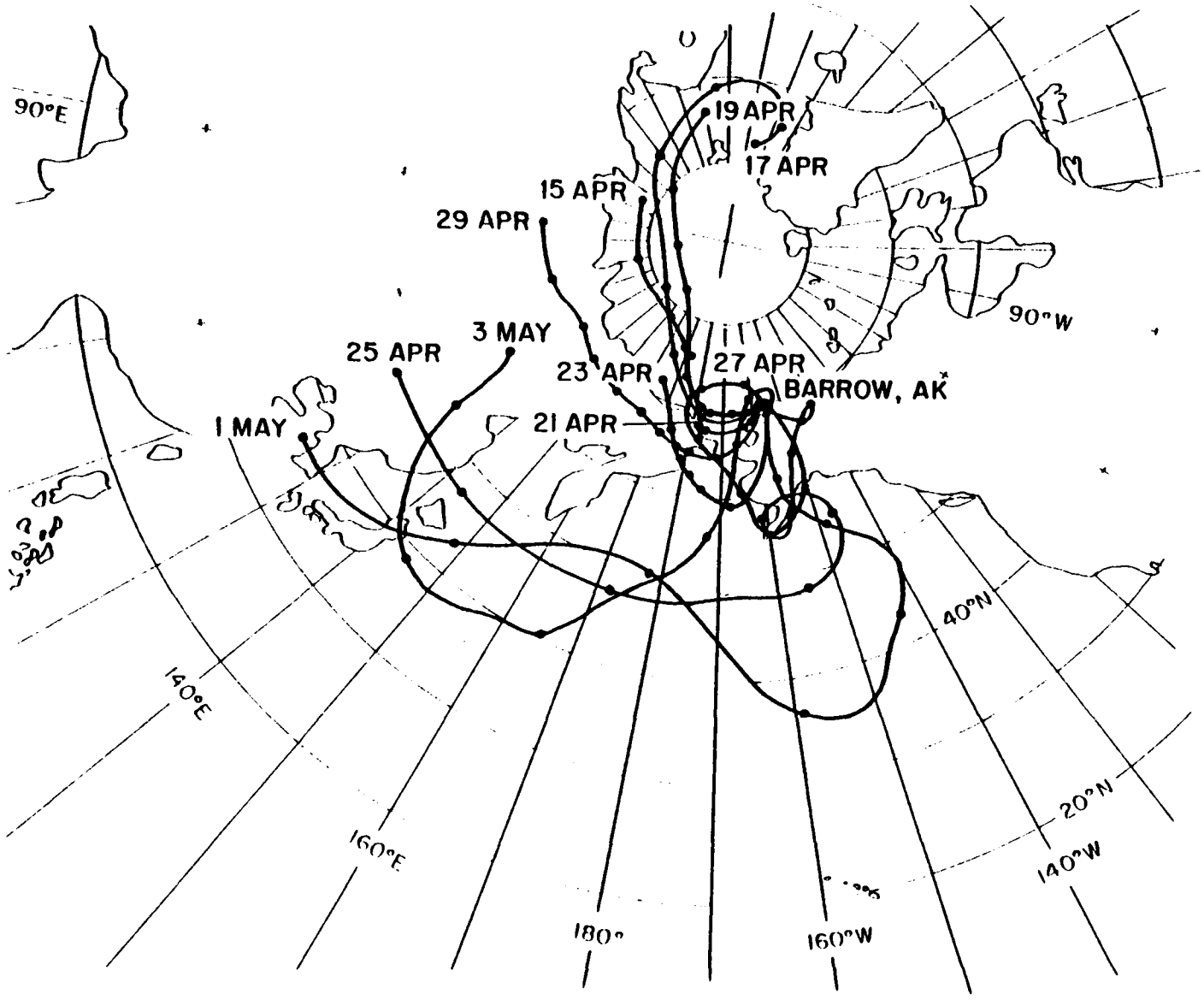


Figure 13. 700-mb isobaric trajectories of air to Barrow. Numbers refer to date of arrival at Barrow, 1976. Solid circles each 24 hours along trajectory.

in the northeast United States and/or Europe. To our knowledge these data are some of the first strong indications of air pollution in the Arctic during winter. Other data which we have taken at the surface near Barrow have revealed that this pollution is routinely present during the entire winter half-year, and, while not high in concentration compared to a city, represents a sizeable percentage of the total Barrow aerosol. A compilation of trace-element data for Arctic aerosol from other locations (Rahn et al., 1977; Section IV. D.3. of this article) has shown that the winter pollution maximum at Barrow is duplicated elsewhere in the Arctic, and probably means that the entire Arctic atmosphere is polluted. The consequences of this pollution are as yet not understood but are potentially serious.

3. Morphology

The conclusion from chemical data that Arctic haze bands were crustal aerosol was confirmed by electron microscopy. Figures 14 and 15 are electron micrographs of typical portions of the Nuclepore filters of filters CD and IJ, respectively. The magnification of the original 10x11 cm negative was 5000X; a convenient reference size for these figures is the Nuclepore pores which are 0.4 μm in diameter. In sample CD many more particles are smaller than the pores than are larger than the pores. The largest particles are few in number and are less than 1 μm in radius. In sample IJ, on the other hand, many more giant particles are seen, with radii up to several μm. Platy layers can easily be seen on several particles; all appear angular and would seem to be crust-derived.

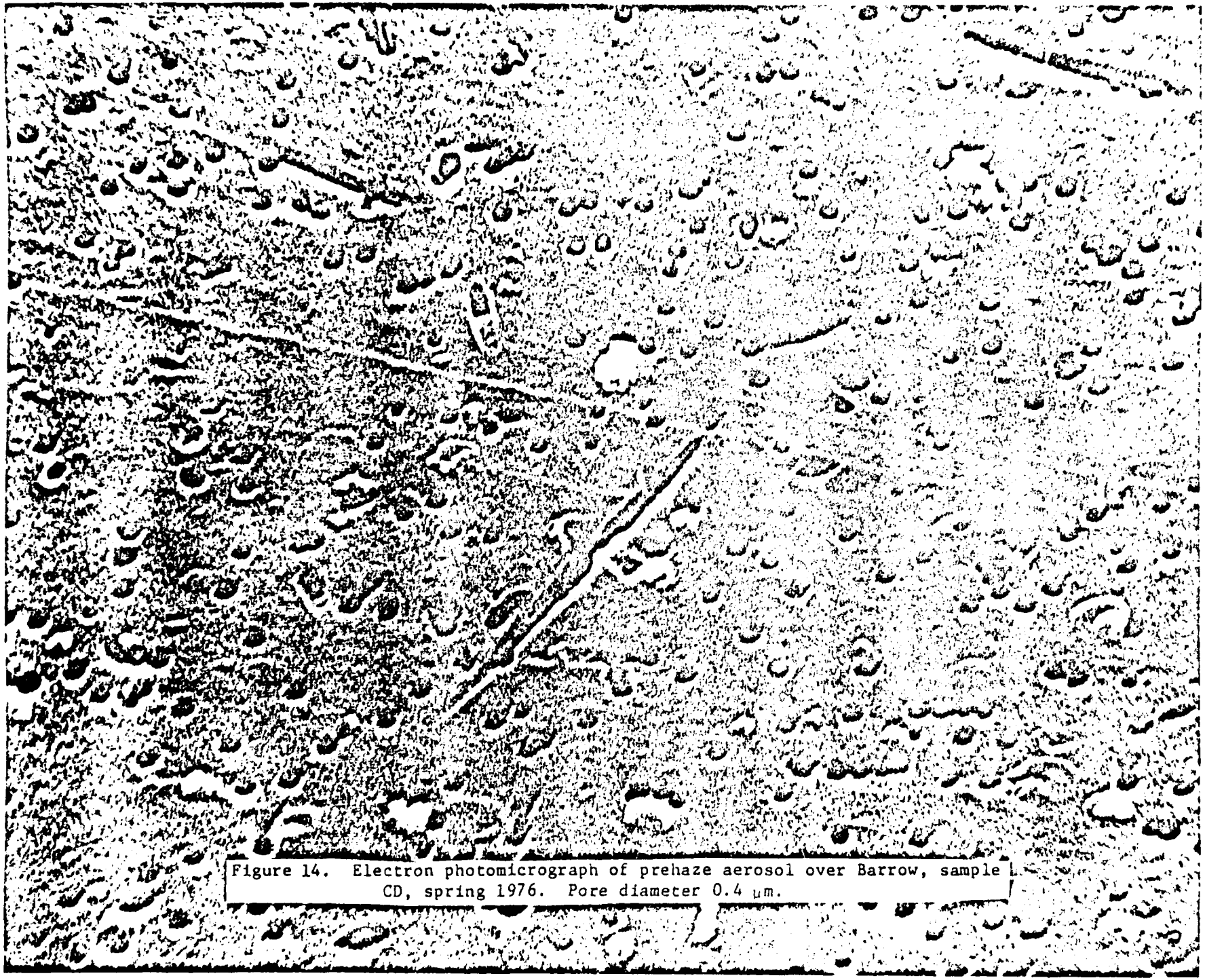


Figure 14. Electron photomicrograph of prehaze aerosol over Barrow, sample CD, spring 1976. Pore diameter 0.4 μm.

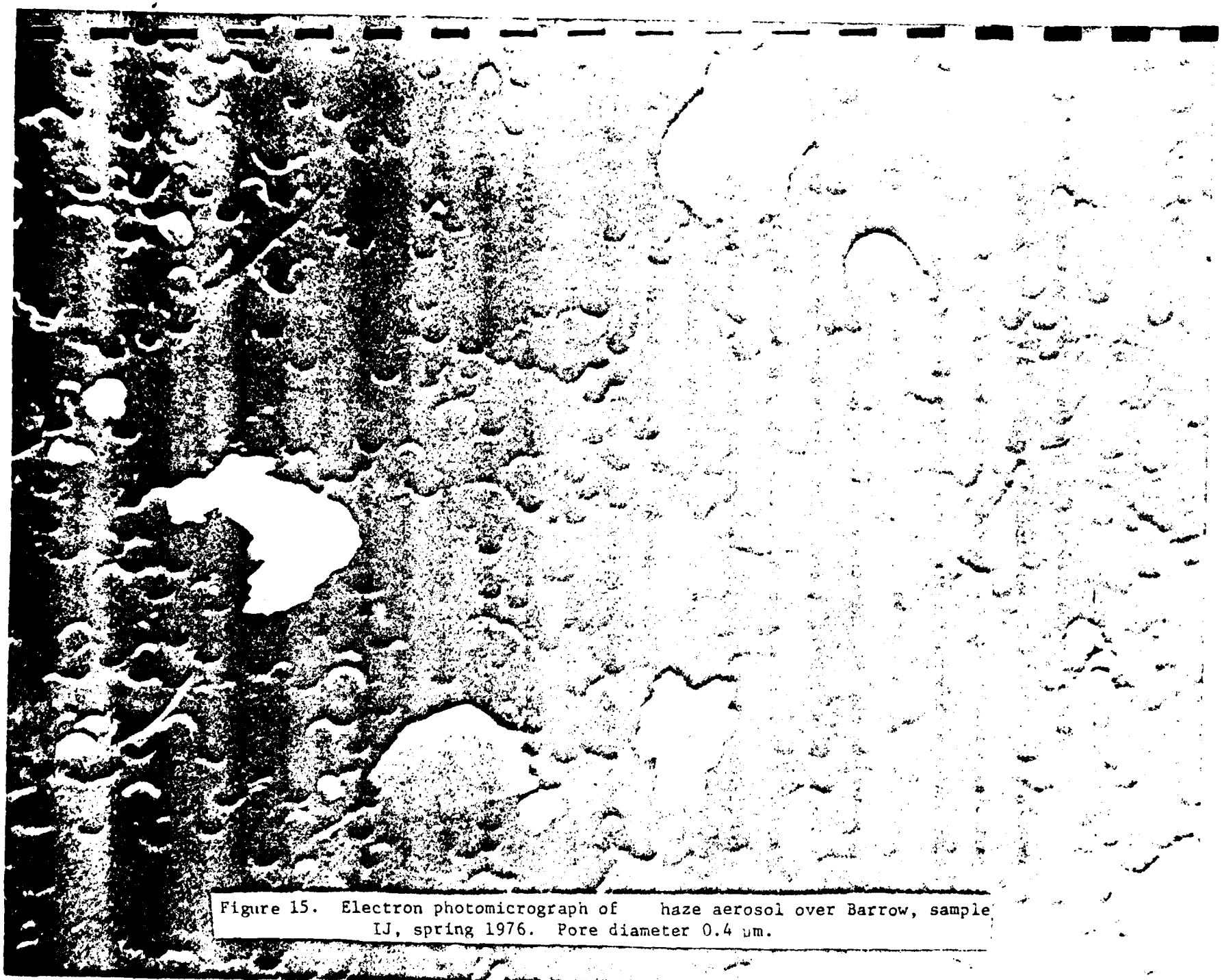


Figure 15. Electron photomicrograph of haze aerosol over Barrow, sample IJ, spring 1976. Pore diameter 0.4 μm.

4. Single-particle analysis

Electron microprobe analysis of individual haze particles offered further evidence of their crustal origin. The larger, angular particles characteristic of the haze layers were all rich in Si, the best indicator element for the crust. In some cases the particles were nearly pure Si, i.e., probably quartz, SiO_2. In most cases high Al accompanied the high Si, indicative of any number of crustal minerals such as clays, micas, feldspars, etc. Si/Al ratios were typically in the range of 1 to 3, similar to the average values of about 2 compiled by Rahn (1976a). Some of the larger particles had high K contents, with K, Si, and Al in ratios near to those of potassium feldspar. In other cases Fe and Mg were high, suggestive of biotite. Occasionally a giant particle would be found which was rich in Ca and S and little else, which reminds one of gypsum, which is known to occur in deserts.

The smaller particles, those with radii less than 1 μm, appeared completely different, being smoothly spherical or hemispherical as opposed to the more angular larger crustal particles. Additionally, the smaller particles differed in elemental composition from the larger ones - the latter were invariably rich in S, the logical explanation for which was droplets of ammonium sulfate. Indeed, under the first pass of the intense electron beam of the microprobe these tiny particles evaporated, and could not be detected at all on subsequent passes. Ammonium sulfate is known to volatilize under these conditions.

5. Size and volume distributions

The particle-size distributions of nonhaze and haze aerosol over

Barrow can be used to gain additional information about the sources and history of the aerosol there. Size distributions for the aerosol of samples E and KL were constructed by manual counting of particles from the Nuclepore filter samples of these flights. Ten randomly taken photomicrographs of each filter were counted at each of two magnifications, 1000X and 10,000X, in order to accumulate adequate statistics. Size classes of a factor of 2 in radius were used, extending from radius 0.025 μm to radius 8 μm. Actual numbers of particles counted per size range varied from a maximum of more than 700 in the smallest range down to 0 to 2 in the largest range. The size distributions for flights E, nonhaze, and KL, haze, are shown in Figure 16. Above radius 1 μm in both samples there were essentially only mineral particles; in the size range just below 1 μm where mineral and nonmineral particles could both be counted, the mineral component is shown as solid circles. Mineral particles were distinguished from nonmineral particles by a crude and perhaps inaccurate method: the mineral particles gave brighter images than the nonmineral particles, and appeared to be the only particles visible under the 1000X magnification. Size distributions derived from these photographs were therefore considered to be exclusively mineral. This procedure may have missed some of the finest mineral particles, but as will be seen below seems not to have missed much of the mineral mass.

The size distribution for flight E seemed to be composed of two reasonably well-defined components, with break point at radius 1 μm. Above this radius the slope of the distribution was very steep, approximately -5. Below 1 μm the slope was more nearly -2, and extended over more than a decade in radius. Absolute values of the ordinate, i.e., the

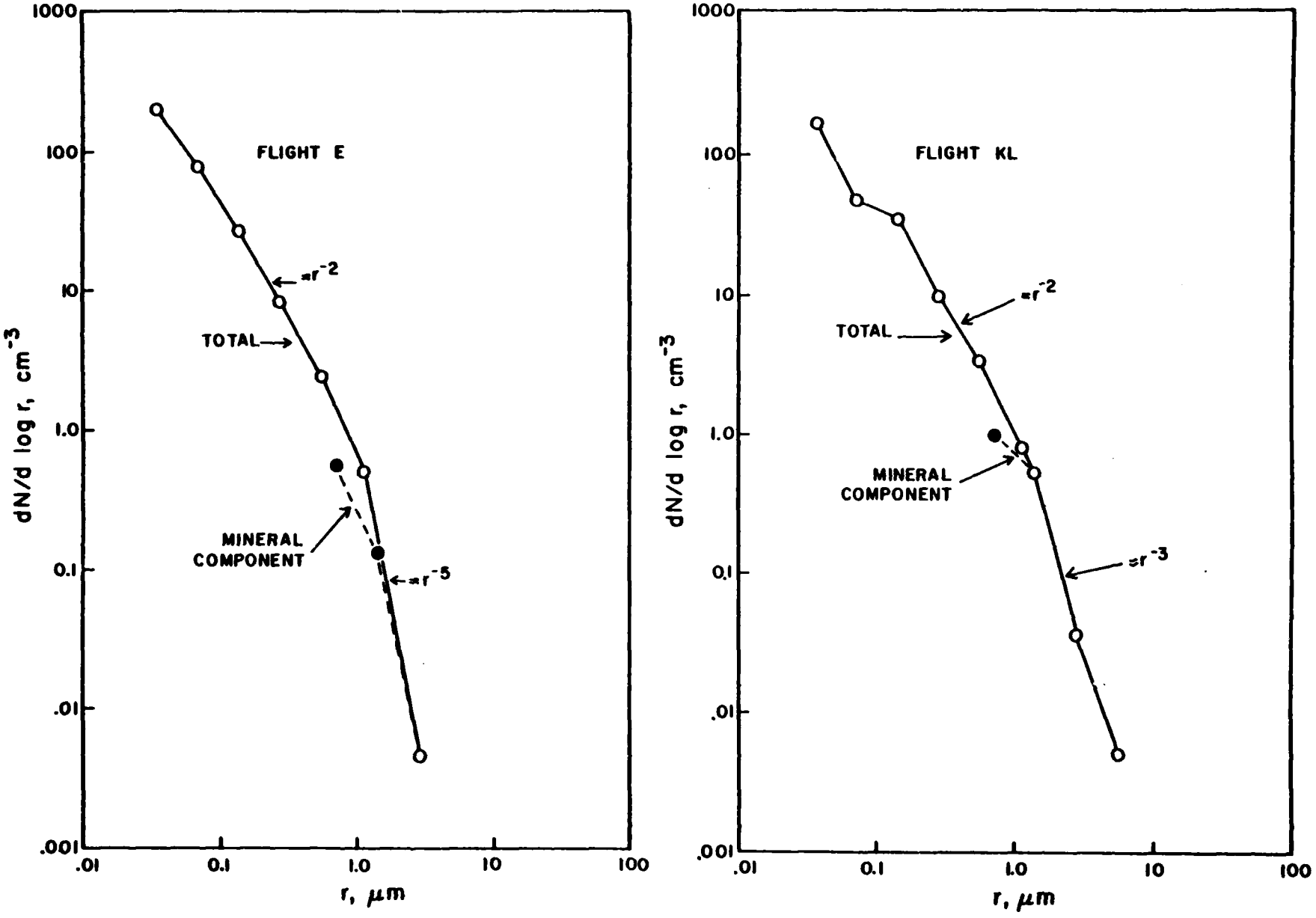


Figure 16. Particle-size distributions for typical prehaze (E) and haze (KL) aerosols.

position of the curve, are typical for background aerosol. The area under the curve, i.e., the total particle concentration for sample E, is about 96 per cc, which is somewhat lower than indicated by the condensation nuclei counter during the flight (this may be because these concentrations are right on the detection limit of the Gardner counter used), but which is nevertheless reasonable for background aerosol (Junge, 1963). Sample KL showed the same two-component distribution, also with break point about 1 μm. The slope below this point was -2 again, but now the slope above 1 μm was more nearly -3, and extended to larger particle sizes. This difference in slope confirmed the visual observation from the Nuclepore photomicrographs of larger concentrations of giant particles in the haze aerosol compared to the nonhaze aerosol. Total particle density in this sample was 87 per cc, essentially the same as for sample E. This shows that even though layers of Arctic haze have considerably different optical properties than nonhaze air, and even though the mass concentration in such a cloud is significantly greater than outside the cloud, the total number of particles involved is still determined by the nonhaze component, which is nearly unaffected by the presence of the haze.

These number distributions have been converted to volume distributions, by multiplying the number of particles within each size interval by $4/3\ \pi\ r_g^3$ where r_g is the geometric mean radius of the interval. The results for the two samples are shown in Figure 17, where it can clearly be seen how the extra volume, or mass, of the haze aerosol was found nearly exclusively in the giant particle range, i.e., with particles or radius greater than 1 μm. In Figure 18 these volume distributions are presented

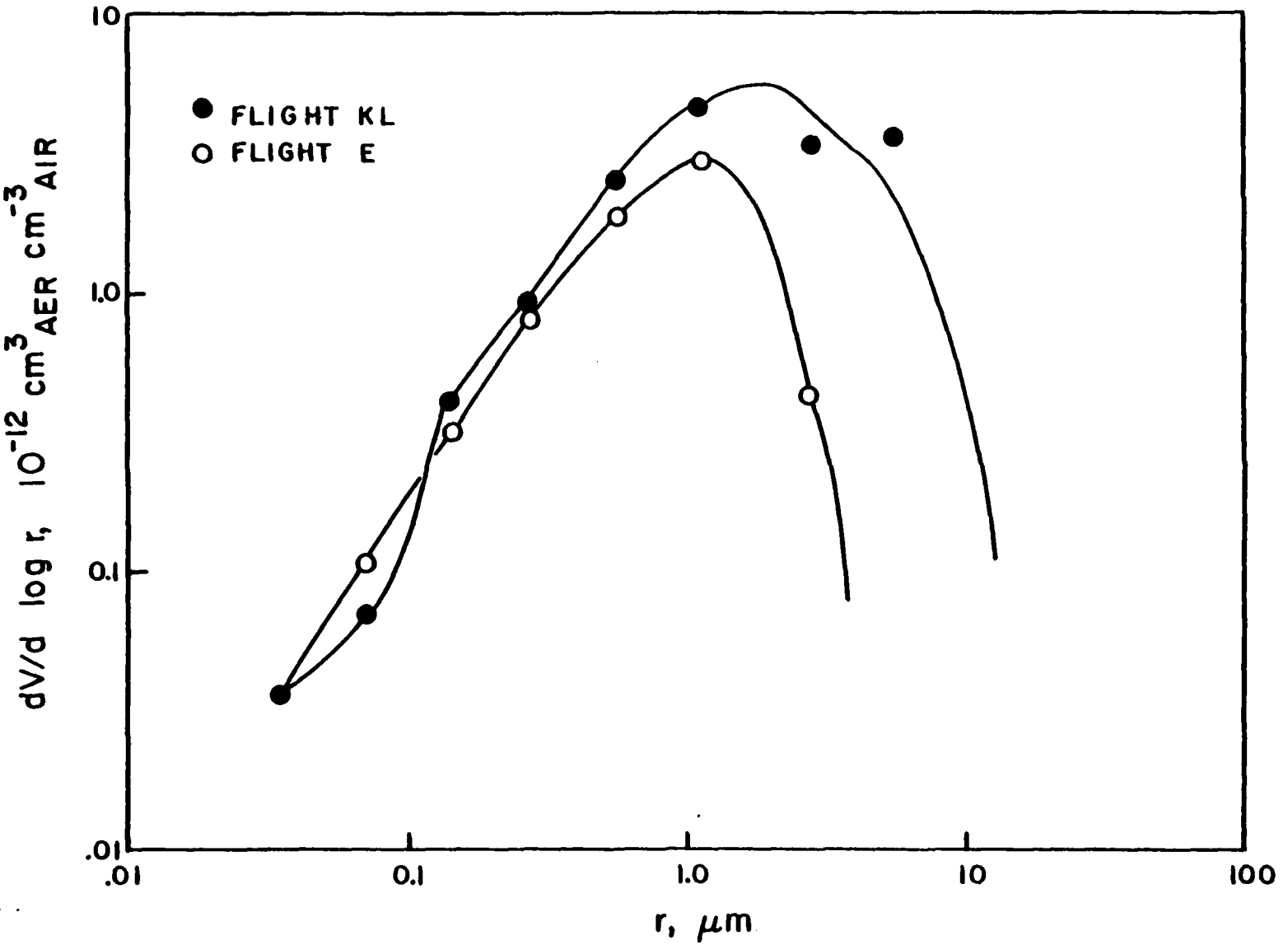


Figure 17. Volume distributions for samples E and KL, Barrow, spring 1976.

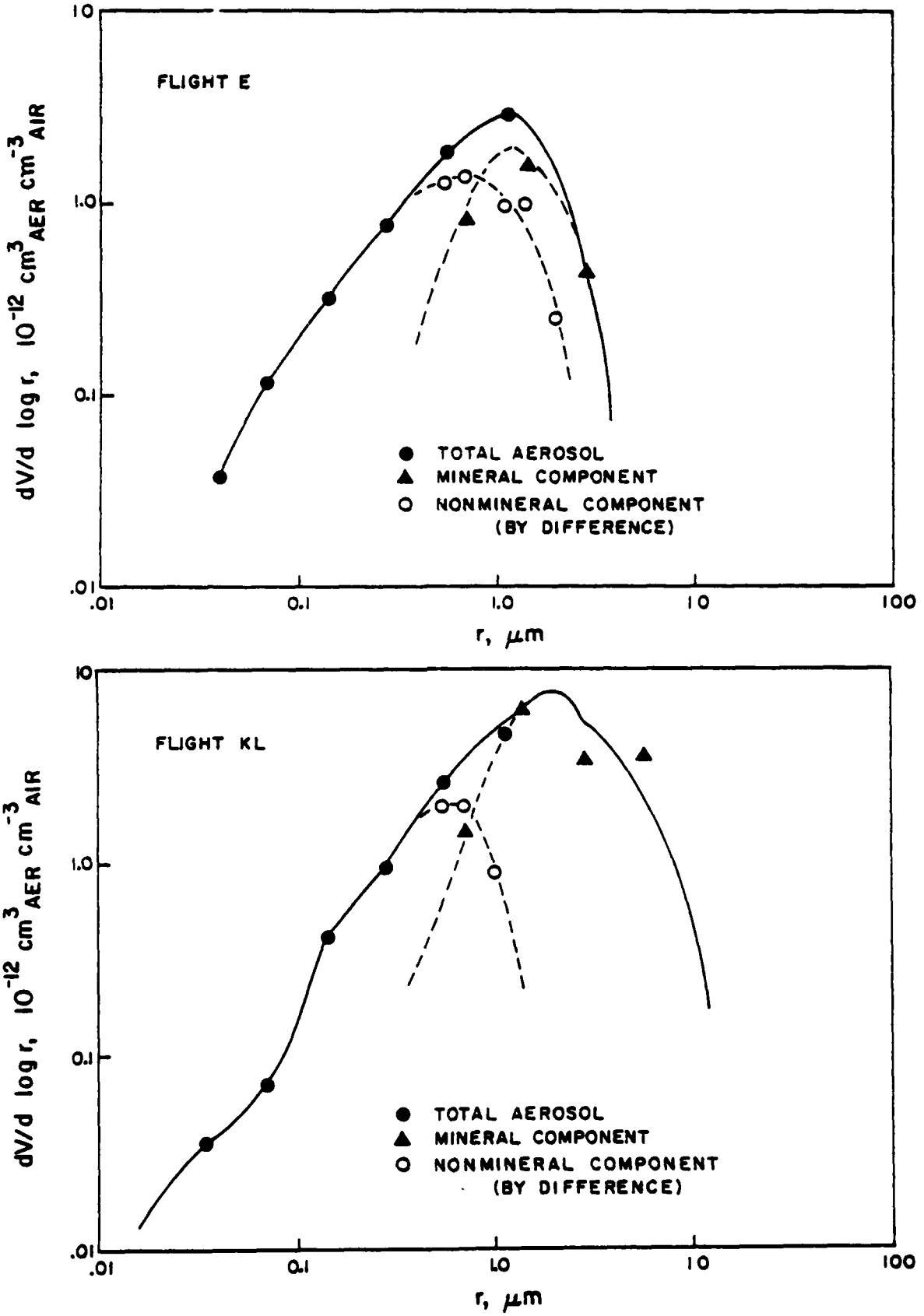


Figure 18. Volume distributions for individual samples E and KL, resolved into mineral and nonmineral components.

for each sample individually, with a resolution into mineral and nonmineral components, the nonmineral component being the difference between the mineral and component and the total aerosol. It appears from these plots that the nonmineral component was probably a single species, because it seemed to have nearly a unimodal log-normal distribution. It did, however, appear to be slightly skewed toward the small-particle end. It is not clear whether this is significant. A two-component aerosol would agree with the visual impression from the electron photomicrographs (Figures 14 and 15). The small-particle component had a mass-median radius (MMR) of approximately 0.6 μm in both samples, which agreed very well with values of 0.4 to 0.5 μm quoted by Gravenhorst (1975) for non-sea salt or "excess" sulfate in marine areas. In contrast, sulfate over middle Europe has been reported to have mass-median radii somewhat smaller, about 0.2 to 0.3 μm (Georgii et al., 1971). Thus the nonmarine aerosol component is in the right size range to be sulfate. Recall from above that it is also composed of S-rich particles which volatilize under the microprobe beam.

The mineral component of flight E appeared to have a narrower but also log-normal volume distribution, with an area or mass approximately the same as for the nonmineral component. In sample KL, on the other hand, the volume distribution of the mineral component was both higher and broader than in flight E, whereas that of the nonmineral component remained nearly unchanged. Note at the same time that the MMR of the mineral component shifted to about 2 μm from about 1 μm in sample E. This demonstrated that the mineral aerosol of the haze was younger than that of the background aerosol.

A clue to the age or distance of travel of the mineral aerosol came from its MMR in Figure 18. Schütz et al. (1978), in a companion paper in this volume, show both by model and by actual measurements that as Sahara dust is transported farther from its source its MMR shifts systematically to smaller sizes. Immediately over the source the MMR is 10 to 20 μm or greater; after 1000 km of transport the MMR is 5 to 10 μm, and by 5000 km from the source, the limit of their calculations and measurements, the calculated MMR had decreased further to about 2 μm. Measurements at this point, however, showed a broader distribution than was expected and a somewhat higher MMR, perhaps because of coagulation during repeated cycles of condensation and evaporation. Our haze band of sample KL also had its MMR at about 2 μm with a surprisingly broad distribution. On this basis we can postulate that the Arctic haze aerosol of at least sample KL was about one week old, the time interval required to transport Sahara aerosol the 5000 km. Because our mean velocity of transport was probably higher than the relatively slow 6 to 8 m sec^{-1} of the northeast trades which carry the Sahara dust, our haze aerosol had probably travelled more than 5000 km. In fact, actual Asia-Alaska trajectories seem to be 12,000 to 15,000 km.

D. Discussion

1. Relationship to previous observations

The Asian source for Arctic haze explains or confirms many previous observations such as: (1) The large horizontal dimension of the haze bands; (2) Prior haze events at Barrow (For example, the episode of March-April 1974 was preceded by a few days of strong flow from

the Asian deserts); (3) Prior measurements of anomalous turbidities over Fairbanks, Alaska (For example, during 17-22 February 1976 there was a whitish haze episode in Fairbanks which affected air at all levels and for which no explanation could be found. Visibilities were reduced from the normal 150 km to less than 30 km. Trajectory analysis revealed that this air was also of Asian origin. Similar haze incidents with trajectories leading back to Asia seem to be a regular feature of the Fairbanks atmosphere during winter and spring.); (4) The accumulation of brownish insoluble deposits in the pack ice north of Barrow, which under examination with a light microscope appear to be continental dust (R. Paquette, personal communication). The mineralogy and possible sources of this dust have also been discussed (Darby et al., 1974); (5) Anomalous ice-nucleus concentrations at College, Alaska; Blue Glacier, Washington; and Nagoya, Japan during February and March, 1968, for which trajectory analysis showed eastern Asia to be the probable source (Isono et al., 1971); (6) A case of abnormally high condensation nucleus concentrations at Barrow in March 1970 (Radke et al., 1976), which was explained as pollution from Prudhoe Bay to the east but which may have originated from Asia because the large-scale flow for this period was from the general direction of Asia; (7) Quartz and illite distribution patterns in deep-sea sediments of the Pacific. Non-biologically produced quartz is found most concentrated in Pacific sediments along a rather narrow line which stretches east-west at approximately 30 to 40° north from Japan to California (Heath et al., 1977). The band follows a remarkably straight track across the Pacific. Its southern boundary is well-bounde , but it- orthern boundary is more diffuse.

There are two indications of lobes to the north, which may correspond to frequent transport paths of Asian dust to the north. One of these is quite near the North American coast, and is placed about the same position as the sharp northward sector of the trajectory of 1 May shown in Figure 13. Illite shows a rather wide band of high concentrations in Pacific sediments, fanning out from the Asian mainland at about 30^{o} north and continuing eastward at this latitude nearly to North America (Griffin et al., 1968). This pattern has been recognized as eolian in nature, and also shows a distinct northward lobe just south of western Alaska, at about the place where some of our dust trajectories turn sharply northwards; (8) The deposition of large amounts of crustal dust in Japan, especially during winter. Ishizuka (1972) reported that winter snows in Japan contained crustal material from the Gobi Desert. Fukuda and Tsunogai (1975) also reported that the Japan Islands were efficient collectors of this crustal aerosol, especially in winter; (9) Eolian additions of quartz to Hawaiian soils. Rex et al. (1969) have shown that Hawaiian soils contain measurable amounts of the marker mineral quartz that could not have originated within the soils themselves. They considered that this quartz must have been derived from the atmosphere because its concentration in soils increases with elevation. They presumed that precipitation scavenging of the atmospheric layer just above the trade-wind inversion accounts for this increase of quartz with elevation. They also noted a general increase in the $^{18}O/^{16}O$ isotopic ratio in quartz of Pacific deep-sea sediments with increasing proximity to the Asian coast, which corresponds to larger particle sizes of this quartz (Clayton et al., 1972). This implies that

the quartz of the Pacific sediments, and probably also that of the Hawaiian soils, has had an Asian origin.

2. Mass flux over Alaska

The mass of desert dust transported into the Arctic seems to be very great. For the five-day episode over Fairbanks discussed above, columnar mass loadings derived from radiation measurements showed values of about 45 mg m^{-2} (for an aerosol density of 2.5). A dust cloud the width of Alaska (900 km) travelling at 80 km hr^{-1} would carry 4000 tons of dust into the Arctic per hour, or a half-million tons over the five-day episode. This is equivalent to skimming off a 0.2-μm layer from a 10^{o}x10^{o} desert. Admittedly these estimates are rather rough, and subject to considerable uncertainty. They do seem, however, to make the point that impressive masses of Asian crustal aerosol routinely reach Alaska and the Arctic.

Clearly then, Asian dust must be considered a major contributor to the overall aerosol burden of the Arctic troposphere. It completely alters the aerosol chemistry of that region when it enters, and affects the radiation balance as well. Further study of this strong source region and the transport of its aerosol to the Arctic is essential to establish the extent of possible climatic effects which it may exert on that fragile region. For example, do dry years in Asia significantly increase the dust burden of the Arctic, and if so, does this significantly alter the radiation balance there? A similar question can be posed concerning the possible effect of changes in the hemispheric circulation to more meridional character, such as occurred during the winter of

1976-77, when Alaska was bathed in warm air from the south while the eastern United States was covered with Arctic air.

But the importance of Asian dust in the atmosphere does not end with the Arctic. Although no quantitative studies are presently available as to the total amounts of crustal aerosol produced annually by the Takla Makan and Gobi Deserts and the Loess Plateau, our rough estimates presented above show that they must be great indeed. Most of this desert dust, the coarsest fractions, is probably deposited within China as loess. Of that which is fine enough to be carried long distances within the atmosphere, the majority travels with the prevailing westerlies and is ultimately deposited in the Pacific Ocean as sediments. Along the way, however, it stays in the atmosphere long enough to affect radiation and precipitation processes. No measurements have been made on these effects. Eventually some of the dust reaches North America, probably anywhere from California to British Columbia. We know of no measurements of Asian dust in these regions. Occasionally the atmospheric flow patterns become temporarily warped, and the dust plume curves northward to Alaska. But this is only a small percentage of the time - the subject of this paper is therefore only a small perturbation of a much larger and more significant phemonenon, systematic study of which remains for the future.

3. The Turbidity Problem

The Asian dust affected the optical properties of the Alaskan aerosol. Because the dust mass was mainly in the giant size range ($r > 1$ μm), one might expect that these particles would have increased

the turbidity mostly at longer wavelengths, which is what was observed. Figure 19 shows the frequency dependence of aerosol optical depths for two typical examples of prehaze and haze periods. In the absence of haze bands the optical depths decrease smoothly by a factor of two between 400 and 900 nm, whereas in the presence of haze bands the dropoff is only 20%.

Based on everything that has been said here so far, one would expect the total turbidity of the atmosphere to be increased significantly by the presence of the haze bands. In fact, the turbidities decreased during our haze episode. This discovery caused us to reexamine our entire view of the Asian dust/Arctic haze relationship. The turbidity data and our interpretation of it is presented here as the concluding section of this paper.

Figure 20 shows the course of the episode as indicated by the Al concentrations, the aerosol optical depth of the atmosphere at 500 nm as measured at ground level, and the temperature at the 500-mb (5500 m) level over Barrow as read from the 500-mb hemispheric maps prepared by the United States National Weather Service. It is striking how the turbidity remained constant while the Al concentration increased by a factor of 20 at the beginning of the episode. Turbidity measurements alone would have missed the entire haze episode. As the episode progressed, however, the turbidity slowly, then more rapidly decreased, until after the episode it reached its lowest level of the measurement period. Before the episode, the turbidity of about 0.2 was typical of the unusually high values previously reported for Barrow in spring (Shaw, 1975). The post-episode value of about 0.07 was much nearer to what one

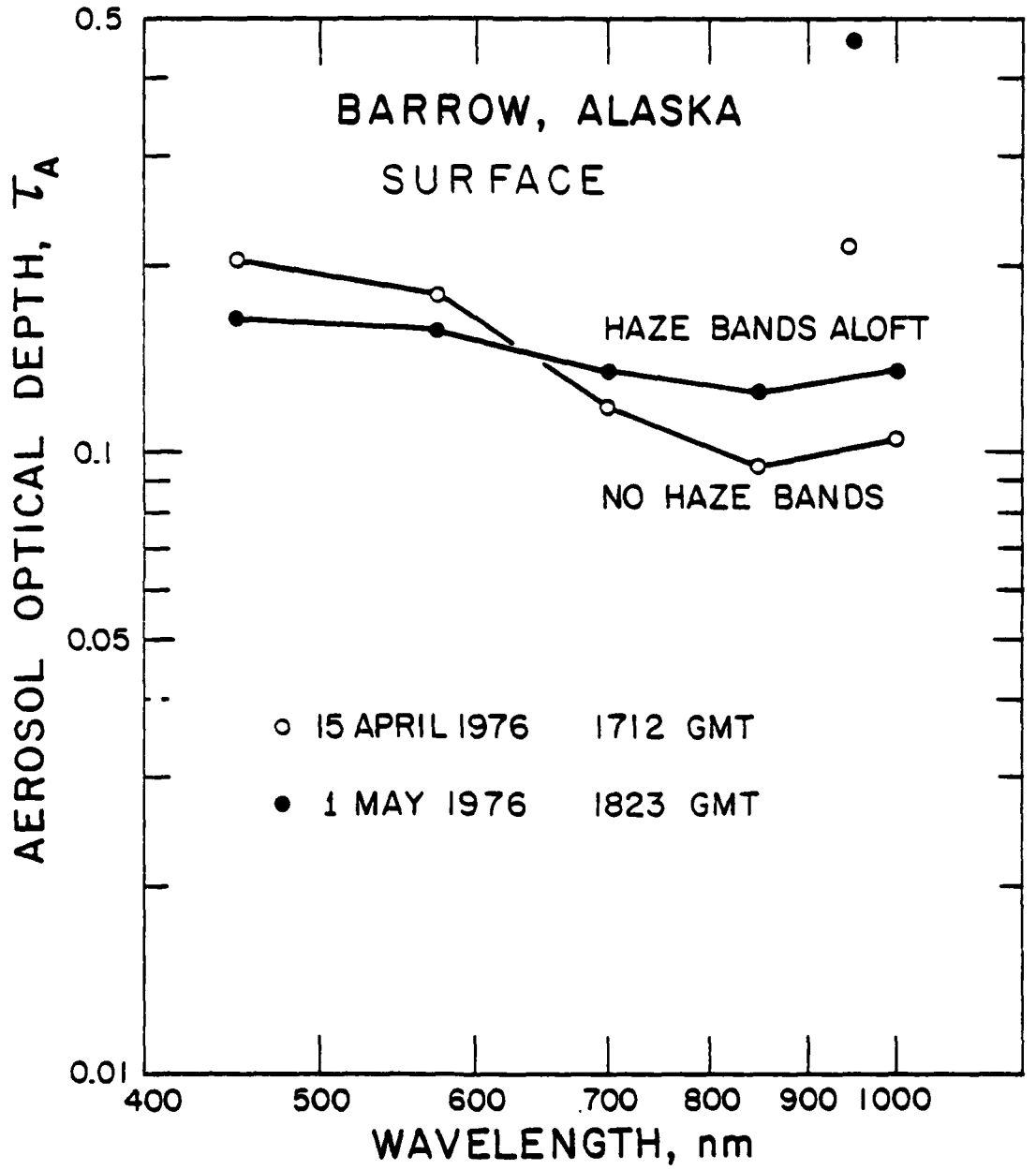


Figure 19. Wavelength dependence of optical depth, nonhaze and haze conditions, Barrow, 1976.

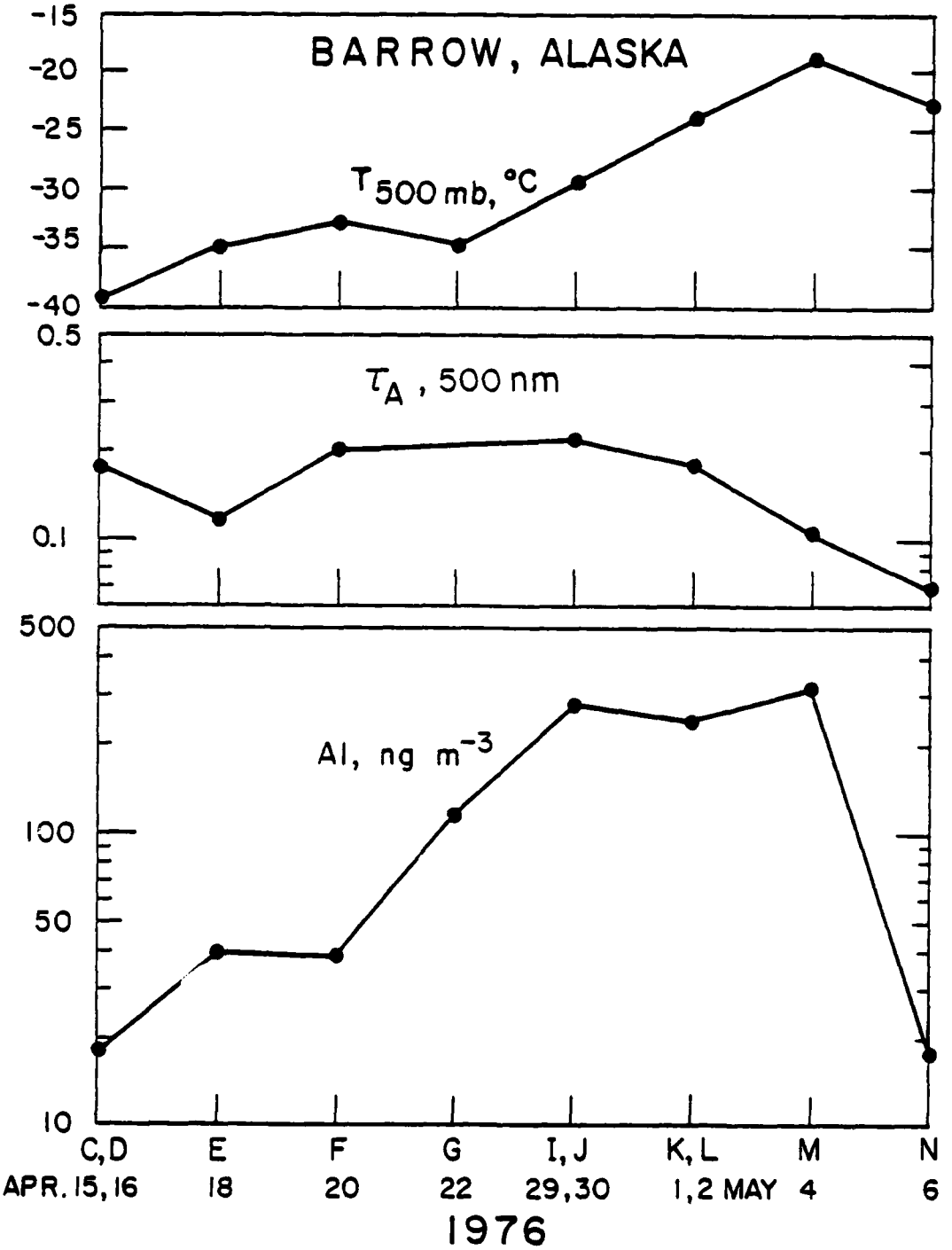


Figure 20. Progression of Al concentration, optical depth at 500 nm, and 500-mb temperature through the haze episode, Barrow, spring 1976.

might expect for "clean" air at Barrow, being about the same as values measured there in summer and fall.

Why then did the haze layers decrease the turbidity instead of increasing it? A clue comes from the 500-mb temperature, which began a steady increase soon after the arrival of the haze, at just about the same time that the turbidity began to decrease. Only at the conclusion of the event did the temperature aloft decrease again. The temperature increased considerably during the episode: 16°C at the 500-mb level and enough at the surface to cause the first local thaw of the season. This temperature rise suggests that air from the south was progressively advected over Barrow during the episode; Figures 21 through 23 are our attempt to illustrate just how massive this advection was, and to demonstrate that during the episode the northern air normally over Barrow was entirely replaced by the quite different southern air.

Figures 21 through 23 contain a series of 18 daily 500-mb hemispheric maps for the period before, during, and after the episode. On each map the Loess Plateau, Alaska, and a few reference land masses are outlined, and the -25C isotherm is darkened. At this time of the year this isotherm can be used as a measure of the approximate center of the 500-mb polar front, north of which is found cold, dry air and south of which is found much warmer, more humid air. In the vicinity of the front temperature, pressure, etc., change more rapidly than they do farther from the jet. In other words, the polar front is a region of sharp contrast where two quite different air masses meet. It is our thesis that if the trend of turbidity over the course of our episode is to be understood, it must

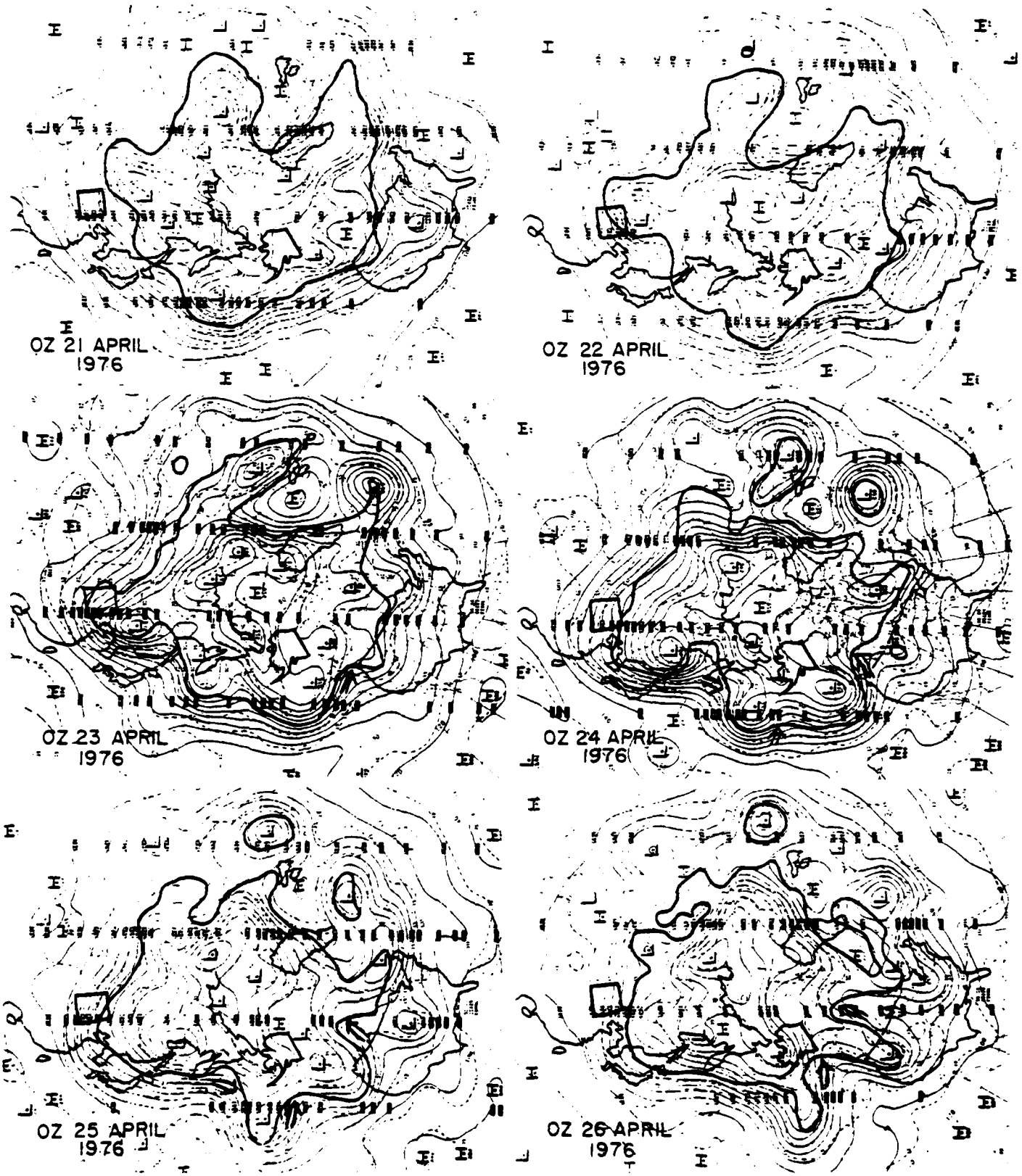


Figure 21. 500-mb contour maps with darkened - 25C isotherm, 21-26 April 1976.

Figure 22. 500-mb contour maps with darkened - 25C isotherm, 27 April - 2 May 1976.

Figure 23. 500-mb contour maps with darkened - 25C isotherm, 3-8 May 1976.

be assumed that these northern and southern air masses also differ strongly in their inherent turbidities, with the northern air mass being significantly more turbid than the southern air mass.

Consider now Figures 21 through 23. At the beginning of the period, around the 21st of April, Barrow was still in northern air, but with weak flow from the south. By 24 April strong waves were developing in the frontal flow near Alaska, with increasing southerly flow over all of Alaska. The -25C isotherm was still well below Alaska, however. Starting on 25 April this isotherm began to move rapidly to the north, and by 30 April it had reached the vicinity of Barrow. On 3 May it reached 82°North, its northernmost extent. At this time the northern border of the front was nearly at the North Pole. One day later, 4 May, the air over Barrow was at its warmest of the period and the dust episode was near its end. At this time air over central Illinois was colder than air over Arctic Alaska.

Note how clearly the tongue of southern air can be followed from these maps as it worked its way northward over Alaska. First signs of the tongue appeared on 25 April. It then developed rapidly, broadening as it built northwards. Because of the massive scale of this outbreak, Barrow and indeed the rest of Alaska were essentially totally in southern air from 30 April through about 5 May. These large-scale intrusions of southern air into the Far North are common. As part of the normal north-south exchange of air they occur throughout the winter half-year, when the north-south temperature contrasts are the greatest, and can be seen on any series of upper-level maps.

Our present conception is that air during April and May 1976

consisted of three zones: turbid air to the north, a polar-frontal boundary which from time to time contained Asian dust, and less turbid, i.e., cleaner air to the south. But why should air to the north be dirtier than air to the south? One usually imagines the opposite. We propose to explain this by developing two hypotheses: that the high turbidity of the North is pollution-derived, and that the concentration of this Arctic air pollution is controlled by hemispheric-scale movements of the polar air/polar front system.

Let us begin with the second, or circulational hypothesis, which can be stated as follows: The polar front separates the two principal air masses of the Northern Hemisphere from each other, and can be considered as a barrier to mixing between them. (The barrier is, of course, not absolute.) Consequently, each air mass is quite uniform within itself but different from the other. To emphasize this simplicity we refer to them as "northern" and "southern" air, respectively. In winter the polar front lies south of major air-pollution sources like the northeast United States and Europe; consequently their effluents mix freely within the northern air mass and pollute it greatly. In summer, this front lies north of the northeast United States and Europe; their pollution then mixes primarily within southern air so that northern air is much less polluted.

This hypothesis is supported by a number of observations. (1) Air pollution of distant origin is routinely detectable in the North, with abundances which can be related to strong sources far to the south. This is documented in Figure 24, a plot of monthly mean concentrations of excess (noncrustal) vanadium for two mid-latitude source areas and

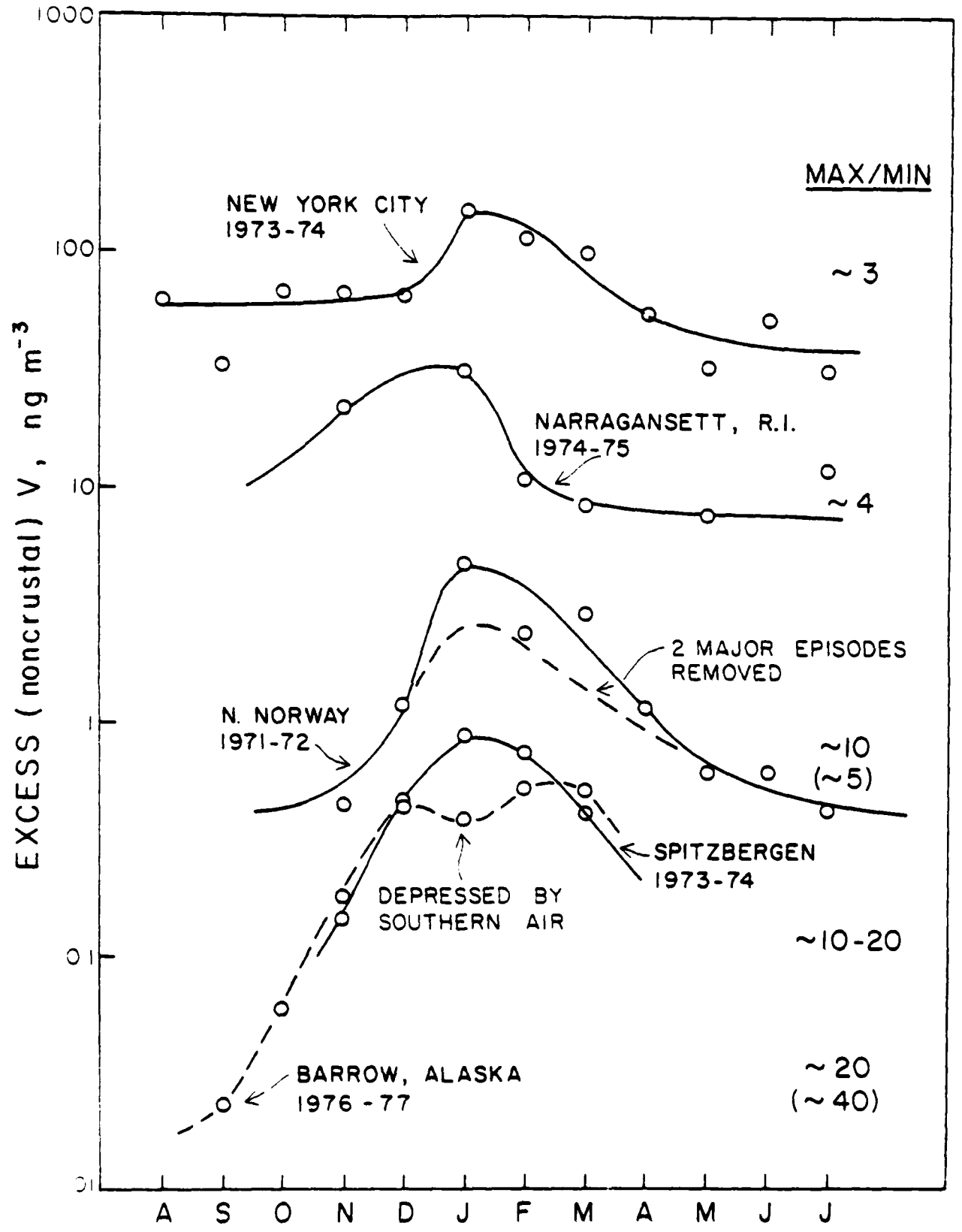


Figure 24. Monthly mean concentrations of excess vanadium at New York City (Eisenbud and Kneip, 1975), Narragansett, Rhode Island (W.F. Graham, personal communication), Skoganvarre, Norway (K. Rahn, unpublished data), Spitzbergen (S. Larssen, personal communication), and Barrow, Alaska (this work).

three remote Arctic sites. In the northeastern United States source area, represented by New York City (Eisenbud and Kneip, 1975) and Narragansett, Rhode Island (Graham, personal communication), excess vanadium shows a peak concentration in January, with non-winter concentrations about three to four times lower. In Skoganvarre (Rahn, unpublished data), northern Norway, January is also the time of maximum concentration, but pollution vanadium is detectable throughout the year; the maximum/minimum ratio is higher than in the source areas, however, being five to 10 instead of three to four. In Spitzbergen (Larssen, personal communication) pollution vanadium is also present year-round and has its maximum in January; the maximum/minimum ratio can only be estimated at 10 to 20 because the record is incomplete. At Barrow the record is also incomplete, but what there is shows a pattern nearly identical to Spitzbergen except for a temporary depression in January and February of 1977 when unusually large amounts of southern air reached Alaska. The maximum/minimum ratio is 20 to 40.

There are several noteworthy regularities of Figure 24. All five locations have winter maxima; all except Barrow peak in January, and Barrow clearly would have a January maximum were it not for the intrusions of southern air (which were unusually frequent in January 1977). With increasing distance from source areas the vanadium concentrations decrease, but the maximum/minimum ratios also increase. These two effects are most pronounced for Norway, Spitzbergen, and Barrow, all three of which are not source areas for pollution vanadium (No. 6 residual fuel oil is not burned in northern areas because its viscosity is great enough for it to require heating during the winter). The fact that the

vanadium maxima of these remote areas occur at the same time as those in the source areas suggest that remote-area vanadium is derived rather directly from source-area vanadium. The increase of maximum/minimum ratios with distance from the source suggests that circulation to these areas in winter is more direct than in summer, which agrees well with the concept of the polar front as a barrier developed above. In summer the barrier is between the sources and the Arctic; in the winter they are both on the same side of the barrier.

From Figure 24 one gets the distinct impression that in winter the entire northern half of the northern hemisphere is a single circulational unit, within which the air pollution emitted near its southern borders mixes freely. As a result, the whole area is polluted. The extreme similarity of vanadium concentrations at Barrow and Spitzbergen, on opposite sides of the Arctic, attests to the surprising uniformity of at least the Arctic part of the area. The Alaskan side of the Arctic is a bit anomalous, being subject to periodic intrusions of dust-laden Asian air throughout the winter, but otherwise is similar to the rest of the Arctic.

(2) A second observation supporting the circulational hypothesis is that the high levels of winter pollution vanadium at Barrow appeared suddenly, about two weeks after the polar front moved to the south. This is shown in Figure 25, a plot of excess vanadium in the Barrow surface atmosphere week by week from September 1976 to January 1977. These data come from a series of high-volume filters taken at the clean-air monitoring site of the Geophysical Monitoring for Climatic Change program of the National Oceanic and Atmospheric Administration, just outside

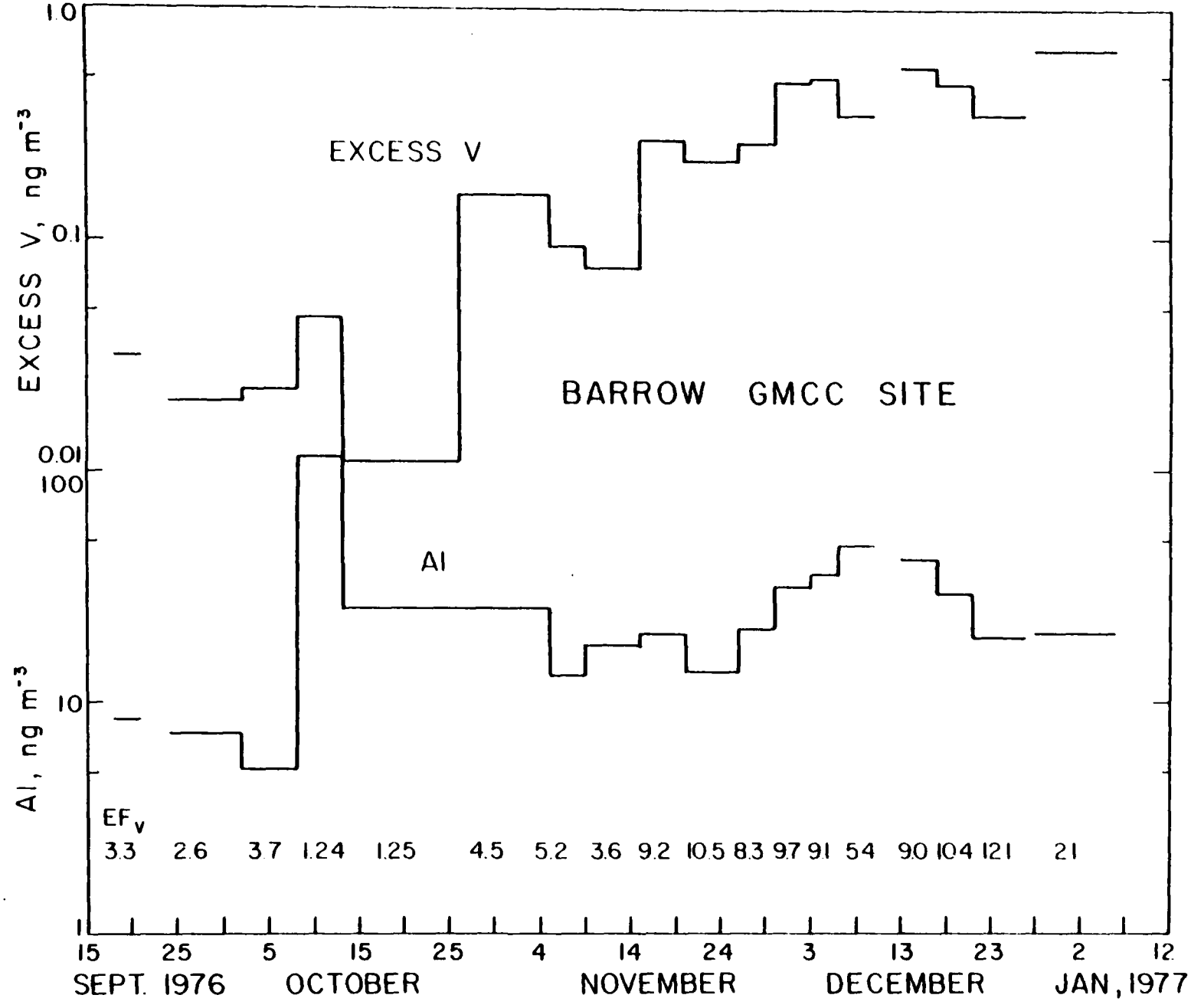


Figure 25. Concentrations of excess (noncrustal) vanadium and total aluminum at Barrow, September 1976 - January 1977.

of Barrow. In September and October, excess vanadium had concentrations of roughly 0.03 ng m^{-3} and enrichment factors of total vanadium were one to four. Then abruptly, about 1 November, the concentration of excess vanadium jumped up by about an order of magnitude, to about 0.2 ng m^{-3}. From November to January, it climbed higher, apparently via a series of pulses. There was no systematic increase of the crustal reference element aluminum from October through January. Thus, as the vanadium concentration increased, so did its enrichment factor, shown at the bottom of the figure. A maximum enrichment factor of 21 for vanadium was seen during the first week of January.

The abrupt ten-fold increase in pollution vanadium about 1 November was completely unexpected and without precedent in our experience. To understand its causes one must think globally. The first of November is only about two weeks after the time when the polar front migrates rapidly southward over North America and other parts of the globe, establishing the winter circulation pattern (Yeh et al., 1959). In 1976 the winter pattern first set in over the northeast United States about the 15th of October, as evidenced by the latitude of the center of the 500-mb front at the longitude of New York City, which is plotted in Figure 26. In the last week of September the front suddenly moved far southward from its summer position, then returned to north of 50° before it settled down to about 40° during October. After October there was little systematic change of its position through the beginning of January.

As of mid-October the northeast United States was therefore in northern air, so that its pollution effluent could now migrate freely to

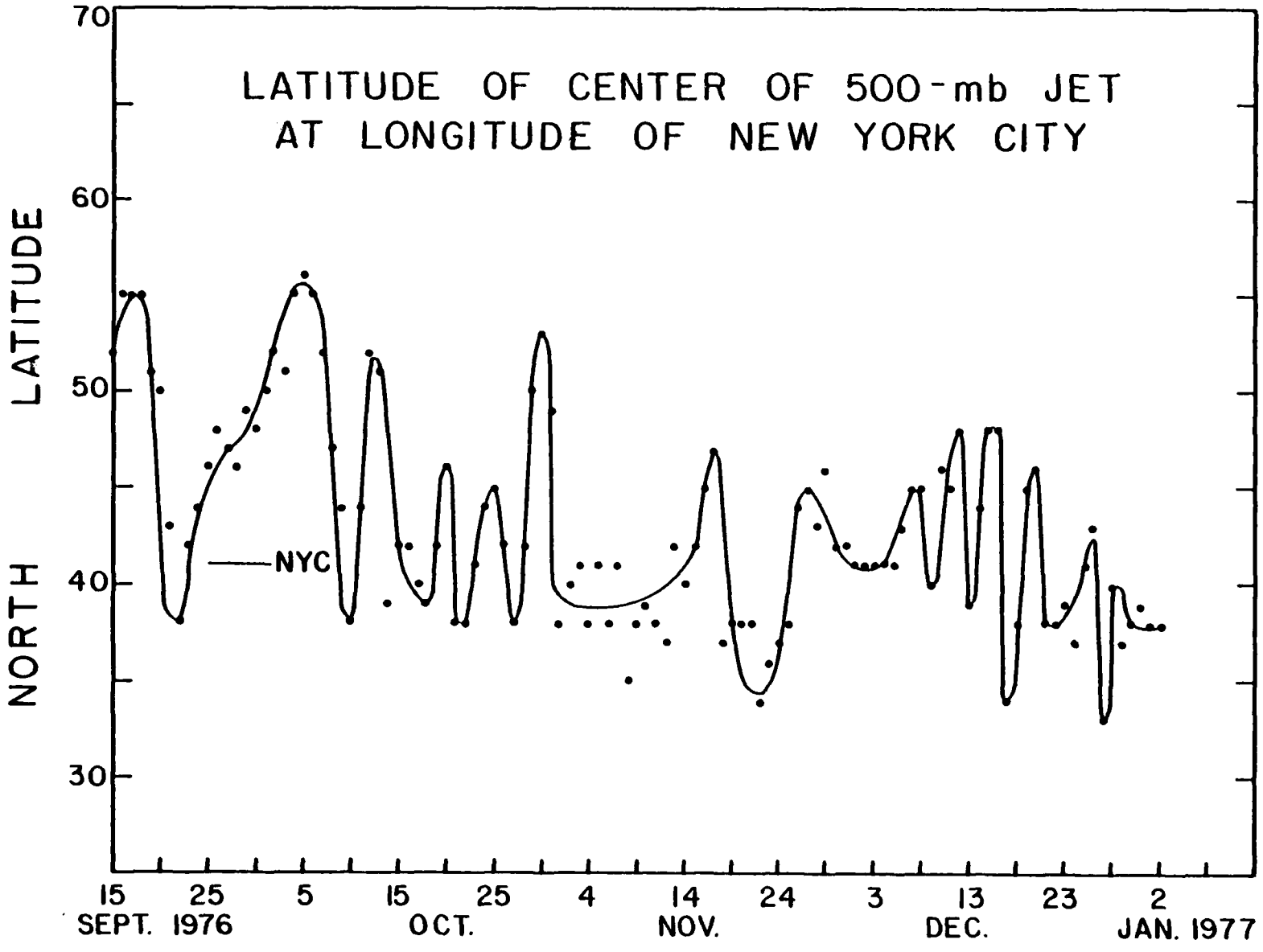


Figure 26. Latitude of the center of the 500-mb polar front at the longitude of New York City, September 1976 - January 1977.

the north. The first pulse of its pollution may have formed a "pollution front," which after a travel time of five to 10 days ought to have been detectable anywhere in the air mass. This timing is not incompatible with the November 1st increase actually seen at Barrow. Elevated pollution levels would be expected to continue in the North until May or June when the polar front recedes to the north (Yeh et al., 1959), as shown for the year 1975-76 in Figure 27. At the time of this writing the vanadium levels were indeed persisting into at least March.

The northeast United States is, of course, not the only source of air pollution for the North, perhaps not even the major one. We used it here as an example because weather data were more readily available for it than for Europe. Europe itself is the other obvious pollution source to be considered, but it seems to be in and out of northern air much more irregularly during the year than does the northeast United States.

This hypothesis of control of pollution levels in the north by the motion of the polar front is admittedly an oversimplification, especially concerning the assumption of pollution homogeneity within northern and southern air masses. We feel, however, that it is a useful starting point, and will attempt to refine it in the future.

We now turn to the first hypothesis, the identification of the high turbidities of Arctic air with air pollution. We developed this idea because of several features of Arctic turbidity which seemed compatible only with air pollution. First, the high turbidity seems to be Arctic-wide, as evidenced by data from Barrow (Shaw, 1975; this work), McCall Glacier in northeastern Alaska (Shaw and Wendler, 1972), and

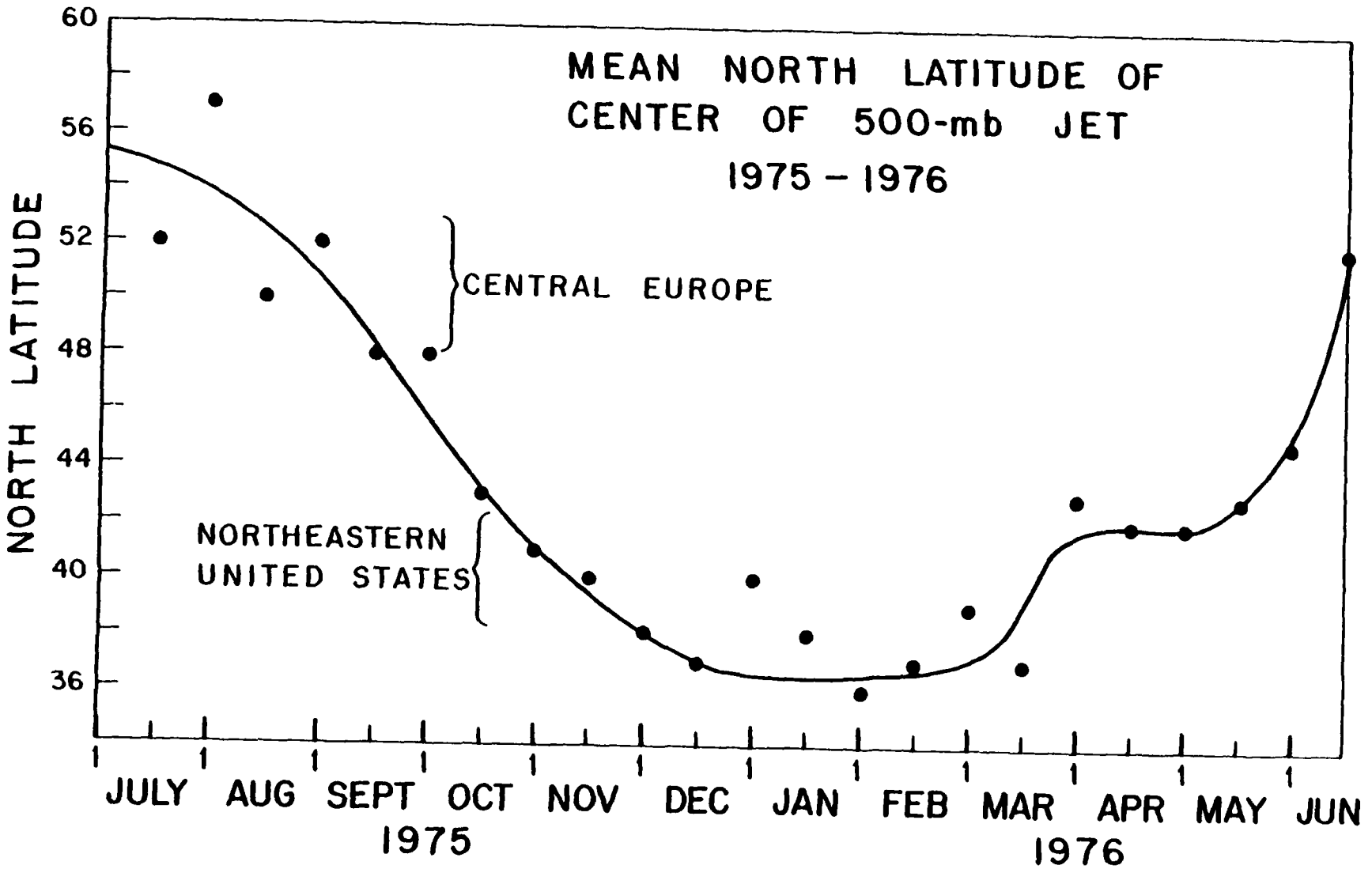


Figure 27. Globally-averaged mean latitude of the center of the 500-mb polar front 1975-1976. Data points twice monthly.

Devon Island in the Canadian eastern Arctic (Holmgren, 1971). We now know, from the data illustrated in Figure 24, that air pollution as well is found over widely separated regions of the Arctic and therefore probably over the entire Arctic. Second, these high turbidities seem to be a spring phenomenon both at Barrow and McCall. This is illustrated in Figure 28, which shows that spring turbidities at both locations are well above the summer values. The summer decrease at Barrow does not show up here as strongly as it probably is, however, because of the small number of data points available. Recent visual observations at Barrow indicate that the atmosphere there was clearer during July, 1977, than March 1977. As we have seen from Figures 24 and 25, air pollution in the North has a winter maximum and a summer minimum. Is this in agreement with the spring maximum of turbidity for the North? We think it may well be, because turbidity measurements need a visible solar disk and so cannot be performed in the Arctic between approximately mid-November and mid-February. Thus, an observed spring maximum of turbidity may be consistent with an actual winter maximum; turbidity would then have the same seasonal pattern as does air pollution.

The third feature of Arctic turbidity compatible with air pollution is its decrease from the Arctic to the Subarctic during winter. Figure 29 shows that average spring turbidities at Barrow are several times greater than the average annual turbidities at Fairbanks, less than 800 km to the south. Why should the turbidity decrease so rapidly between Barrow and Fairbanks? We had no idea until our ground-level aerosol measurements at Ester Dome, Alaska (just outside of Fairbanks), during fall and winter 1976-77 (concurrent with the Barrow surface

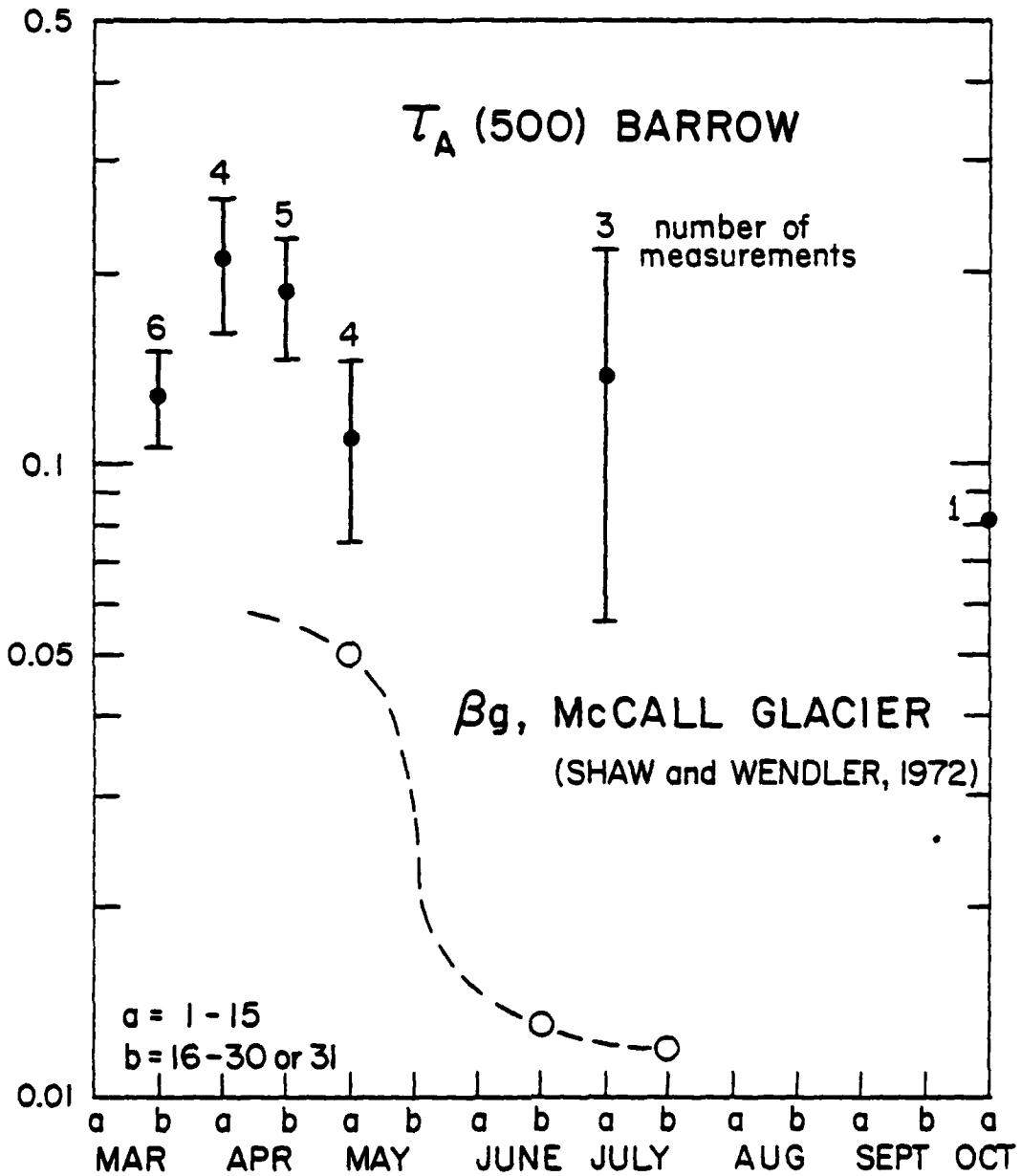


Figure 28. Seasonal variations of turbidity at Barrow and McCall Glacier.

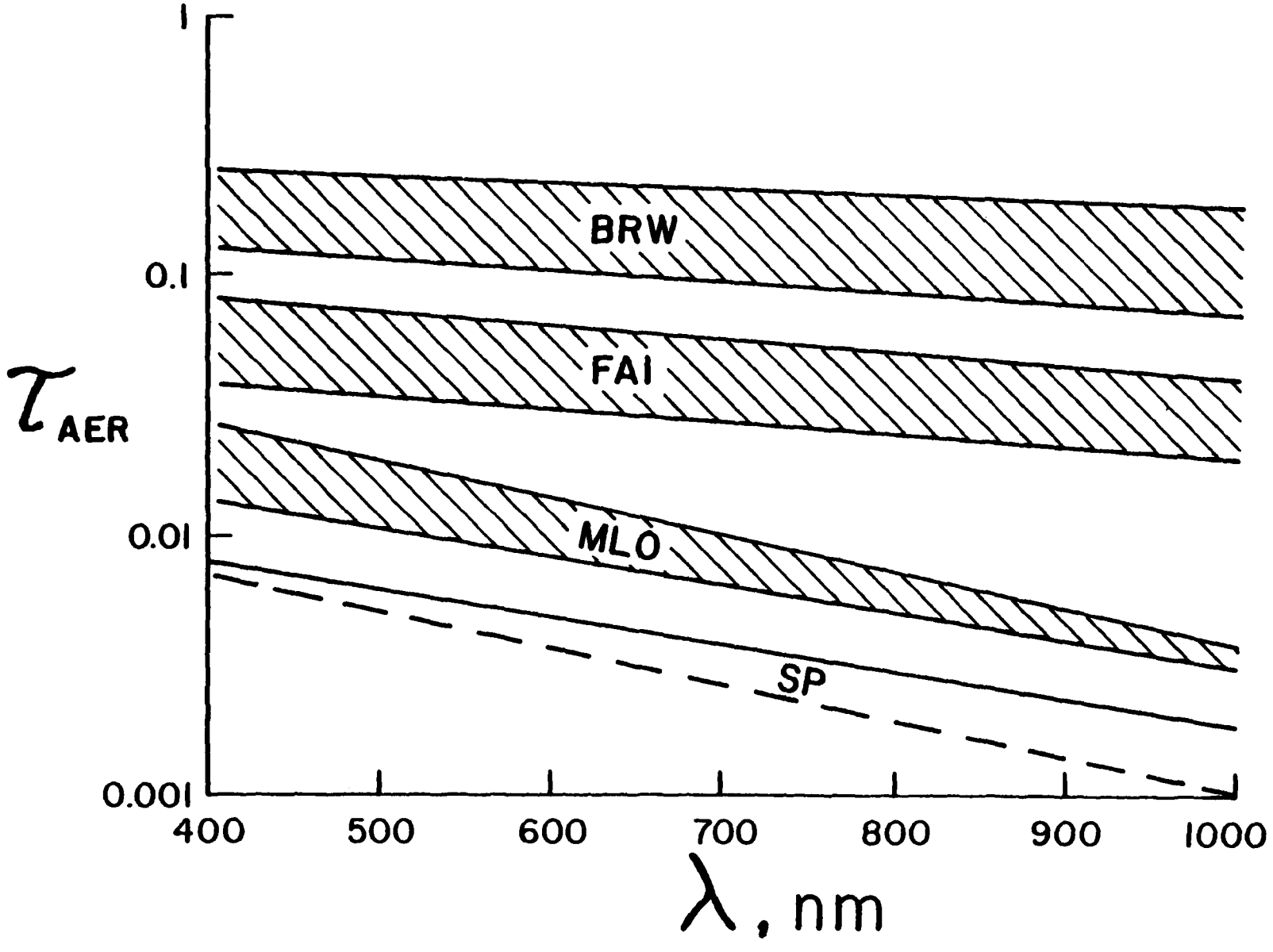


Figure 29. Optical depth vs wavelength at Barrow, Fairbanks, Mauna Loa, and the South Pole.

samples shown in Figure 25) revealed that the large upward jump in several enriched trace metals at Barrow about the first of November was not duplicated at Fairbanks. As soon as we assumed that these metals were pollution-derived at both locations, an explanation took shape: the high pollution levels at Barrow (and the rest of the Arctic) during winter could be due to rather direct air flow from strong sources such as Europe and the northeast United States; three examples of this flow appear as the transpolar trajectories of Figure 13. During winter, however, Fairbanks is shielded from direct penetration of air masses from the north by the Brooks Range - its temperatures are much warmer than at Barrow and its pollution levels are apparently several times lower. Thus the degree of Arctic character of an air mass during winter can apparently be linked with both its turbidity and its air-pollution levels.

In conclusion, then, there seems to be at least two haze-producing agents for Barrow: Asian dust from the southwest and air pollution from the north (ultimately from the south). We now refer to the resultant haze types as type A (for Asian) and type B (for background) Arctic haze. The future is sure to see a great refinement in these ideas; at the moment, though, this dichotomy is convenient.

Out of this study of the Barrow aerosol one thing seems clear: The otherwise clean Arctic atmosphere is not nearly ar far removed from the influence of distant aerosol sources as has been imagined. Far from being truly "clean," the Arctic atmosphere now appears to be a reservoir for large quantities of natural and pollution aerosol, especially during winter. Their effects on Arctic energetics can no longer be neglected. Natural aerosols like Asian dust have been present on a

geological time scale and are thus an inherent part of the Arctic energy balance; pollution aerosol, on the other hand, is much newer, is probably increasing in concentration, and may be considered as a pertubation of the natural state. Possible consequences of this disturbance are treated below and are of greatest concern.

The turbidities of the Arctic atmosphere indicate that approximately 10% of the radiation interacts with the aerosol, whether Asian dust or urban pollution. The direct effect of this interaction is probably a heating of the Arctic atmosphere (Shaw, 1976). How great is this heating, and how might it affect the size of the polar ice cap, weather patterns, etc.? These southern aerosols, whether desert dust or air pollution, are richer in ice nuclei than the usual northern air. Do these ice nuclei affect cloud patterns and consequently modify the precipitation patterns and albedo of the north? If cloud cover were increased by extra nucleiation, would the effect on temperature be a cooling, which would tend to offset the direct warming effect of the aerosols? If so, would the net temperature effect be a heating or cooling ?

If it were a cooling, the possibility of a positive feedback loop might exist whereby pollution cooled cities, cities used more heating, more pollution was generated, the increased pollution cooled the cities further, et cetera ad glacies. While this is only speculation at present, it does seem to illustrate the complex and possibly fragile relationships between the Arctic atmosphere, distant but strong aerosol sources, and possibly even global temperatures.

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E. Science article: Arctic-wide aerosol pollution (14 pp.)

Arctic-wide aerosol pollution

To be submitted to Science, August, 1977

ABSTRACT

Pollution vanadium is routinely present in the Arctic atmosphere. It decreases in concentration with distance from mid-latitude urban areas and exhibits pronounced winter maxima which become less marked near temperate locations. These characteristics are consistent with large-scale aerosol pollution of the Arctic, derived from mid-latitude sources and transported under seasonal constraints of the general circulation.

The Arctic is becoming increasingly developed. Human activity there is now causing local aerosol pollution on a much larger scale than just a few years ago. Development of the North often runs ahead of environmental studies, however, and in most cases proper documentation of the pre-existing or "background" aerosol, necessary to assess the impact of development on Arctic air quality, is not available.

This paper is concerned with the chemical composition of this "background" Arctic aerosol. We present here some recent evidence which strongly suggests, contrary to current opinion, that the "background" Arctic aerosol is substantially pollution-derived, especially in the winter, and that much of this pollution seems attributable to distant mid-latitude sources rather than to sources within the Arctic itself.

We use vanadium (V) as an indicator element for the presence of mid-latitude pollution aerosol. The global strengths for various sources of vanadium have been reviewed elsewhere[1,2], with the conclusions that crustal fragments are the predominant natural source and that combustion of residual (#6) oil is the predominant pollution source. On a global basis crustal sources outweigh pollution sources, but in or near polluted areas this order is reversed. Combustion of heavy residual oil is limited to temperate areas such as the contiguous United States and central Europe, because its high viscosity requires that it be kept warm for ease in handling. Most northern locations, such as Canada and northern Europe, burn lighter grades of fuel (like #2 heating oil) which contain negligible amounts of vanadium[3].

The existence of single major natural and pollution sources for atmospheric vanadium lends it self well to resolving the total vanadium concentration of an aerosol into its pollution and natural components. To do this, we calculate

the crustal component and subtract it from the total vanadium to obtain an excess, or noncrustal, vanadium component, which is then assumed to be pollution-derived:

$$V_{EXCESS} = V_{TOT} - V_{CRUST} = V_{TOT} - Al(V/Al)_{ROCK} \quad (1)$$

Al is the concentration of the crustal reference element aluminum in the aerosol and ROCK refers to average crustal rock[4]. This subtraction is reasonable because in most cases the crustal component of vanadium is only a few percent of the total. The worst case of the data to be discussed below was mid-summer in the high Arctic, where crustal vanadium occasionally reached 50 to 75% of total vanadium.

Strictly speaking, excess vanadium as defined here represents only the pollution component derived from residual oil. Coal combustion, for example, releases pollution vanadium to the atmosphere in fly ash, but because the V/Al ratio of coal fly ash is close to that of the crust[5], this vanadium is treated as natural by equation (1). In most cases, including the Arctic, coal-derived vanadium is only a small part of the total pollution component, however, so that equation (1) remains valid to better than 10%. In any event, the mid-latitude specificity of the residual-oil vanadium makes it a more valuable tracer for Arctic applications than does the less latitude-specific coal-derived vanadium.

We have used equation (1) with recent data to calculate excess vanadium concentrations in aerosols from 5 locations: the mid-latitude source regions of New York City[6] and Narragansett, Rhode Island[7]; the subarctic Skoganvarre, Norway[8]; and the Arctic Spitzbergen[9] and Barrow, Alaska[10]. Monthly mean values

are plotted for these locations in Figure 1, from which a number of regularities are evident:

(1) Excess vanadium is routinely detectable at all 5 locations at all times of the year. Even in the high Arctic all weekly samples showed its presence. This fact is somewhat at variance with the impression obtained from two recent reports of vanadium in the Arctic atmosphere. In one of these, aerosol samples taken on a cruise from Iceland to Nova Scotia showed rapidly decreasing vanadium concentrations and aerosol-crust enrichment factors (defined as $(V/Al)_{AEROSOL}/(V/Al)_{ROCK}$) near Iceland[2]. The crustal or near-crustal enrichment factors in the vicinity of Iceland, coupled with data from the second report for vanadium in Greenland ice cores that also show near-crustal enrichments which have not changed from 1600 AD to the present[11], tempts one to conclude that pollution vanadium is not yet present to any great extent in Arctic air. Our evidence - which is based on longer sampling at more Arctic locations - suggests, however, that such is not the case. On the contrary, pollution aerosol and its associated vanadium would seem to be commonly found over the entire Arctic. We believe that the disagreement between the earlier data and ours is more apparent than real; possible syntheses of these data sets are presented below.

(2) The concentration of excess vanadium decreases with distance from the strong source areas, but seems to be much more nearly constant within the Arctic. From New York to Narragansett (a small Rhode Island town within the general northeast United States source area but itself only a very weak source) the concentration drops off by roughly a factor of 5, approximately the same factor as between Narragansett and northern Norway. The Spitzbergen and Barrow values are another factor of 5 below the northern Norway values. Even

though Spitzbergen and Barrow are on opposite sides of the Arctic, they have nearly identical concentrations of excess vanadium.

From these observations we conclude that mean concentrations over the 5 sites are compatible with vanadium injection in mid-latitude source areas followed by transport, with associated dilution and removal, to the Arctic. Once in the Arctic, however, the dilution and removal processes seem to be much less active, probably because of the extreme stability of the Arctic atmosphere and the relatively slight precipitation, both of which combine to increase aerosol residence times. Thus the Arctic, at least on the time scale of a month or so, would seem to be rather homogeneously polluted.

(3) All 5 sites have a winter maximum of excess vanadium. All except Barrow have a January maximum, and Barrow probably would have also had a January maximum if its Arctic air of winter 1976-1977 had not been displaced by unusually large amounts of southerly Pacific air. (Penetration of a certain amount of southern air over Barrow is, however, a normal feature of the winter circulation there, and among other things this southern circulation brings large amounts of Asian desert dust to Barrow[12]). The overwhelming impression created by these winter maxima is that vanadium in the entire North must be derived rather directly from strong sources far to the south. Thus at least in winter the whole northern half of the Northern Hemisphere contains sizeable amounts of pollution aerosol, so that the term "remote" when applied to this atmosphere has quantitative but not qualitative significance.

(4) The winter/summer, or maximum/minimum concentration ratios for each site also show an increasing trend with distance from the strong sources. From values of 3-4 in the northeast United States they pass through intermediate values of 5-10 in northern Norway (the value depends on whether two strong

episodes of transport of pollution aerosol from central Europe during the first 3 months of 1972 are included), and settle down to values of 10-40 at Spitzbergen and Barrow. Because of the incomplete records at both latter locations, the true winter/summer ratios can only be estimated. As was the case for trends of absolute concentrations, the Arctic is again different: winter/summer ratios are significantly higher there than elsewhere.

Much of Figure 1 can be explained meteorologically. The position of the polar front may be the determining factor for the northern spread of vanadium and other aerosol pollutants[13]. Briefly, the hypothesis is that the polar front forms a sort of barrier or hindrance to meridional transport of polluted air. In winter this front is situated south of the northeast United States and Europe, where much of Northern Hemispheric pollution is emitted. This pollution then circulates freely within this "northern" air mass, and creates the winter vanadium maxima reported here. In summer the front is located to the north of these same sources, so that their pollution is largely cut off from direct entry to the North. As a result, Arctic pollution levels in summer are 10-40 times lower in summer than in winter, and may on occasion be even lower. Nevertheless, Arctic aerosol pollution is still present in summer and can be detected with existing techniques. This summer pollution may be linked more with Europe than with the United States, because Europe's more northerly location, combined with the large meridional summer migrations of the polar front in that area, appear to bring Europe into northern air more often during summer than is the case for the United States.

Interestingly, the transition from summer to winter pollution regimes (at least at Barrow) appears to be quite sudden, occurring in mid-October or early November when the jet stream/polar front system moves rapidly southward to its winter location[13]. It is not yet known whether this sharp increase of

pollution is a general feature of the Arctic atmosphere. It is not observed in subarctic northern Norway, probably because the polar front oscillates over this region all year long.

Of course, the position of the polar front is not the only determinant of pollution transport to the Arctic. Summer/winter precipitation differences are probably also important. From the northeast United States via Iceland and Spitzbergen to the Arctic there are at least 3 precipitation regimes to consider: the northeast United States where precipitation is nearly the same in all seasons, the north Atlantic where precipitation has winter maximum, and the Arctic where it probably has a summer maximum. Because precipitation is so meager in the Arctic at all seasons, the Atlantic effect may be the dominant one. It would tend to decrease the observed winter vanadium maximum in the Arctic. This precipitation effect may be roughly a factor of 2, and so would seem to be small compared to the barrier effect of the polar front, which is shown below to be at least a factor of 30.

This barrier effect of the polar front nicely explains the vanadium data between Iceland and Nova Scotia[2] and shows why this data is not necessarily typical for that region. The Iceland-Nova Scotia samples (University of Rhode Island R/V Trident, cruise TR-102) were taken from 2-24 August 1971, starting from Iceland. The first two samples (2-7 August), where the vanadium enrichment factors were roughly 2 and 4, respectively, were taken close to Iceland when the ship was well north of the polar front. During the last 5 samples (9-24 August), in which the vanadium enrichment factors ranged from 20 to 80, the ship was in the vicinity or south of the polar front, and the air flow was directly from the northeast United States. Across the polar front the excess vanadium dropped in concentration by a factor of 30 to 120,

with a value of 30-50 probably being the most representative. Both the magnitude of this decrease and the actual concentrations are very close to what we have measured from winter to summer in the Arctic: the excess vanadium concentrations for the two samples near Iceland, 0.02 and 0.06 ng m^{-3}, agree well with our September 1976 average of 0.02 to 0.03 ng m^{-3} for Barrow, and the maximum concentrations of 2 to 2.5 ng m^{-3} for the more southerly 5 samples are close to but somewhat above winter means for Sptizbergen and Barrow. On this basis we propose that the concentration profile across the polar front at a given instant in time is analogous to the seasonal profile at Arctic locations, because they are both created by the polar front and its migrations. Thus the Iceland-Nova Scotia vanadium data confirm our ideas about the importance of the polar front in Arctic air chemistry.

The seeming discrepancy between the temporally constant near-crustal vanadium enrichment factors in Greenland ice cores[11] and the dominance of pollution vanadium in the Arctic as revealed by our data is not as easily resolved. It is possible that the Greenland aerosol incorporated into the ice (elevation 2500 m) represents a fundamentally different regime from the surface aerosol of our Arctic sites. If most of the pollution were limited to the near-surface layer below 2500 m and if crustal aerosol were transported preferentially above this height, two quite different aerosols could be created. Layers of concentrated crustal aerosol from the deserts of eastern Asia have indeed been observed over Barrow at altitudes of about 2000 m[12], but it is not known whether these layers reach Greenland in significant numbers. Also, little is known about the vertical distribution of pollution

aerosol over much of the Arctic. Optical measurements of total aerosol over Barrow suggest a scale height of about 1.4 km, which is equivalent to about 75% of the aerosol below 2000 m[14]. This two-regime argument is still speculative, however; much more data is needed before it can be proposed seriously.

The ultimate consequences of large-scale aerosol pollution of the Arctic as proposed here are not yet known. In general, polar aerosol should have a heating effect[15], but this might be partially offset or even reversed if pollution aerosol, acting as a nucleating agent, were to increase the cloud cover and hence the albedo of the North. A nucleation effect could also perturb the precipitation patterns of the Arctic. Precise evaluation of the directions and magnitudes of these and other possible effects must await quantification of pollution aerosol in the Arctic.

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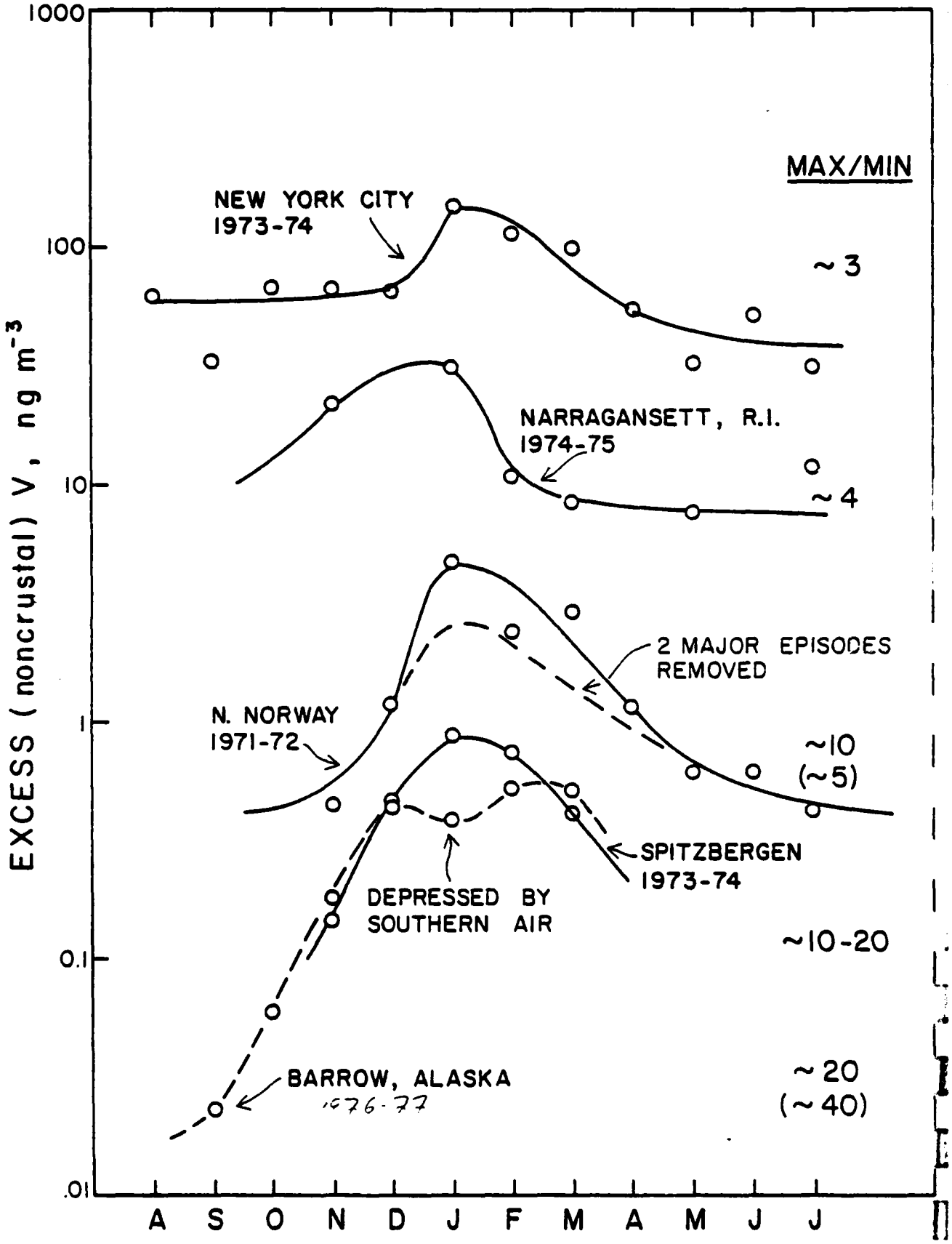
FIGURE CAPTION

Figure 1. Monthly mean concentrations of excess vanadium in the surface atmospheres of 5 mid-latitude, subarctic, and Arctic sites.


1000
100
10
1
0.1
.01
EXCESS (noncrustal) V, ng m^{-3}
MAX/MIN
NEW YORK CITY
1973-74
~3
NARRAGANSETT, R.I.
1974-75
~4
2 MAJOR EPISODES
REMOVED
N. NORWAY
1971-72
~10
(~5)
SPITZBERGEN
1973-74
DEPRESSED BY
SOUTHERN AIR
~10-20
BARROW, ALASKA
1976-77
~20
(~40)
A S O N D J F M A M J J

F. AAAS article: Saharan dust transport over the North Atlantic Ocean (46 pp.)

SAHARAN DUST TRANSPORT OVER THE NORTH ATLANTIC OCEAN

by

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Abstract

The Saharan Desert is one of the major sources of atmospheric mineral dust which is transported long distances. The westward transport is discussed by means of a two-dimensional steady-state transport model and measurements in surface air across the North Atlantic. The model is based on the specific flow conditions for this area and considers sedimentation and turbulent diffusion of dust particles. During preferential transport above the trade wind inversion layer, the aerosol is strongly depleted of particles with $r > 1$ μm within 1000 km from the source. Particles with $0.1\ \mu m \leq r \leq 1.0\ \mu m$ are only inappreciably removed from the dust plume. Actual mineral aerosol mass concentrations and size distributions seem to be approximated best by the model when one assumes a power-law size distribution at the source with $\nu^* = 2$ for particles with $0.1\ \mu m \leq r \leq 20\ \mu m$. Based on this assumption and supported by turbidity measurements, an annual mass budget has been calculated for Sahara dust transported over the North Atlantic for various distances from the source. About 260×10^6 tons of mineral dust per year leave the Sahara westward. Deep-sea sediment data show no basic discrepancy with values predicted by the model.

Resumee

Le desert du Sahara exercise un impact important sur le budget atmosphérique des aérosols minéreaux. Le Sahara est une des sources dominantes de particules minerales transportées à travers l'atmosphère sur de longues distances. A l'exception d'événements ponctuels sur l'Europe, le transport des poussieres sahariennes est bien connu au-dessus de l'Océan Atlantique. Le transport principal a lieu dans la zone des vents alizés de NE où cette poussière se surimpose à l'aérosol maritime et contribue à une certaine fraction des sédiments marins .

Après un exposé des connaissances générales sur les mechanismes de transport, un modèle est présenté qui decrit les conditions de transport à longues distances au-dessus de l'Atlantique. Une attention particulière est apportée aux conditions d'écoulement dans la zone des vents alizés de NE. Les distributions horizontales et verticales sont examinées en fonction de la distance de transport depuis la source. Les distributions volumétriques au niveau de la mer et les concentrations de masse ainsi obtenues concordent avec les données expérimentales obtenues au-dessus de l'Atlantique. Sur la base de mesures d'opacité et du modèle de transport proposé une estimation de l'emmission totale du Sahara est presentée. Finalement la validité du modèle de transport sera démontrée par une comparaison entre les quantités de poussières deposées calculées d'après ce modèle et celles obtenues d'après la literature concernant les sediments marins.

1). Introduction and Survey

Aerosol particles influence atmospheric processes such as extinction of solar radiation and formation of precipitation. They consist of different inorganic and organic materials depending on their origin. One can distinguish in general between two main production processes supplying the atmospheric aerosol. First, mechanical processes at the surface of the continents and the oceans generate particles with $r > 0.1$ μm. Secondly, particles form in the atmosphere itself by gas-to-particle conversion and subsequent coagulation; this process is important for particles with $r < 0.1$ μm.

In this paper we are concerned with mineral particles generated by mechanical weathering such as wind abrasion. Roughly 30% of the surface area of the continents is barren and unprotected from wind erosion. Mineral particles may be injected into the atmosphere from these surfaces.

Approximately 20% of the global production of natural aerosol particles is mineral dust, i.e., soil and rock debris (SMIC, 1971). This amounts to 100-500 x 10^6 tons yr^{-1}. A certain fraction of these particles can undergo long-range transport in the major wind systems, as indicated by their worldwide deposition in the oceans (Griffin et al., 1968) and their contribution to soil formation (Yaalon et al., 1973).

One important source area for mineral dust is the Sahara Desert. Through natural weathering processes at the desert surface large amounts of mineral dust are produced and emitted into the atmosphere. Under suitable flow conditions large amounts of Sahara dust are transported to the Near East, Europe and the Caribbean Sea.

This phenomenon has been well-known for more than one hundred years, since Ehrenberg (1862) published a map (Figure 1) of the global distribution of historical dust-fallout records, with special attention to Sahara dust. For instance, off the West African coast a large dust-fallout area can be seen in Figure 1, called the "Dunkelmeer" (i.e., "dark sea"), which has been recorded since 1160 A.D. by the Arab Edrisi as mentioned in Ehrenberg.

Darwin (1846) reported that in the Cape Verde Islands region the North Star very often disappears at about 30° above the horizon due to high atmospheric turbidity, which led to problems in local navigation. The extent of this dust-fallout region was later described by Semmelhack (1934). According to a long series of observations from ships passing the West African coast, the "dark sea" is found between 10° - 25° N latitude and from the coastline of West Africa to 35° W longitude. Semmelhack also reported a seasonal variation of the frequency of dust fallout, with maxima in January and August, which he explained by variations of the flow conditions in the trade-wind region due to a seasonal shift of the intertropical convergence zone. The winter maximum for dustfall, however, has not been confirmed by other investigators. Semmelhack further claimed that the main transport took place above the trade-wind inversion.

Distinct source areas for dust emerging from West Africa are not well known. Satellite pictures showing dust after heavy sandstorms in the atmosphere cannot be used to determine small-scale source regions, so normally only a few surface observations from weather stations are available. According to a compilation of surface wind and sandstorm observations for the western half of the Sahara from 1925 - 1950 by Dubief (1953), the area of southwest Algeria, Morocco and Mauretania

can be assumed to be a very frequent source region. This idea is supported by a map of atmospheric turbidity for the western Sahara (Figure 2) by Jaenicke (1977), which is based mainly on sunshine recorder data compiled by Dubief (1959). This map can be taken as a rough outline of the source region of dust that can be transported westward.

Sahara dust transport across the North Atlantic was mentioned by Junge (1956). He found reddish-yellow dust in Florida aerosols, the origin of which could be traced on weather maps back to the Sahara Desert. No further work was done on this topic until a study by Delany et al. (1967). They examined airborne dust collected on the island of Barbados. Thorough investigations of the mineral and biological composition of the dust indicated that this matter originates from the African continent and most likely from the Sahara Desert. The size distribution and mineral composition of both the airborne dust and sediments of the subtropical Atlantic were similar.

Delany's work stimulated numerous investigations on the physical and chemical characteristics of Sahara dust over the Atlantic. The mineralogy and chemical composition are discussed in detail by Delany et al. (1967), Folger (1970), Parkin et al. (1970), Chester et al. (1971 a, b, c), Chester et al. (1972), Parkin et al. (1972), Aston et al. (1973), Rahn et al. (1976) and Lepple et al. (1976). Sea-level mineral mass concentrations at the West African coast were found to be 100-10,000 $\mu g\ m^{-3}$ (Lepple, 1975); over the western and middle Atlantic the concentrations were 10-100 $\mu g\ m^{-3}$ (Rahn et al., 1976 and Prospero et al., 1977b). For comparison, in 'clean' maritime air total aerosol mass concentrations amount to only 10-20 $\mu g\ m^{-3}$ (Ketseridis et al., 1976). From these figures, it is clear that these mineral particles contribute to the atmospheric dust burden and to the deep-sea sediments.

Particulate matter dissolved from Sahara aerosol may also influence the chemistry of the ocean. This is indicated by studies of Lepple (1975), Nehring (1976) and Graham et al. (1977), which have shown that a portion of phosphorus present in the Sahara aerosol is released upon contact with sea water. According to Graham et al. (1977), the input of soluble phosphorus near the West African coast from airborne dust is relatively insignificant compared to that from upwelling in the ocean itself. In the nutrient-poor waters farther to the west, however, the atmospheric flux may be of the same order of magnitude as the input to the mixed layer from the deep waters.

Since 1970 more detailed studies on the transport mechanism of Sahara dust crossing the Atlantic were reported by Prospero et al. (1970, a, b; 1972; 1977a), Carlson et al. (1972) and Diaz et al. (1976). These authors investigated the transport by means of vertical wind, temperature, and humidity profiles, aircraft dust and Radon-222 soundings at various distances from the Sahara, and turbidity measurements in surface air. Their conclusions are in general agreement with the rough earlier findings of Semmelhack (1934). Vertical soundings of air conductivity by Gringel et al. (1977) also confirm the specific upper-level Sahara transport layer.

The important features of transport can be summarized as follows: Intensive insolation in the Sahara Desert creates strong surface winds and large-scale convection, which lift dust particles to elevations of approximately 6 km. These dust-laden, dry air masses leave Africa and move westward in the steadily blowing "Harmattan", and reach the Caribbean after one week. During this time the Sahara air masses flow above the trade-wind inversion, which is located about 1.5 km above sea level. The dust layer ranges from about 1 to 5 km elevation near the West African

coast and converges to a vertical layer between about 1.5 and 3 km over the Caribbean Sea. Whereas the lower limit of the dust layer is well defined by the trade-wind inversion, the upper limit is determined by several other factors. These are mainly the vertical extent of the original dust layer over the source area and the balance between vertical motions within the dust layer and above, where subsidence of the subtropical air masses is dominant. The dust layer ranges from about 15°N to 25°N latitude.

Carlson et al. (1972) compared the Sahara dust plume with a "duct flow." During transport, dust particles are removed from the duct by sedimentation and turbulent mixing. Due to the presence of the strong trade-wind inversion, however, which suppresses vertical exchange between the dust layer above and the relatively cool and moist trade-wind boundary layer beneath, one has to expect different removal processes for larger dust particles than for smaller particles. Giant particles (radius $r > 1$ μm) fall out rapidly due to their relatively high sedimentation velocities, and remain practically unaffected by turbulent diffusion. In contrast, particles with $r < 1$ μm, which are mainly subject to turbulent diffusion, will be affected in their downward transport by the trade-wind inversion. Thus giant particles reach the ocean surface within relatively short distances from the source area, whereas the submicron fraction is deposited only after long travel distances.

Air-mass trajectories in the trade-wind boundary layer over the eastern part of the subtropical North Atlantic clearly show that no advection of dust-carrying air masses from the continent takes place. Close to the West African coast surface winds are reported by Emery et al.

(1974) to blow parallel to the coast and at Dakar westerly winds[1] predominate of the year. Thus, the trade-wind boundary layer is assumed to be originally free of dust and to receive its mineral-dust content exclusively from the dust reservoir above the inversion layer.

The seasonal variation of solar radiation causes seasonal changes in both dust generation in the Sahara and transport conditions over the ocean. Superposition of both effects produces a seasonal variation in the large-scale dust transport over the Atlantic Ocean as seen in Figure 3. Considering the sandstorm frequency for different areas of the western half of the Sahara Desert as a measure of its source strength, a seasonal variation of a factor of about two can be seen (according to a compilation of observation data for the years 1925-1950 by Dubief, 1953). Increased sandstorm activity lasts from early spring until fall. The vertical zonal wind field over the Atlantic (according to mean monthly wind data for 1951-1960 at Sal in the Cape Verde Islands by SMN (1968) shows favorable transport conditions in the summer season between May and October. During that time easterly winds prevail throughout levels up to 6 km (500 mb). For the remainder of the year dust transport to the west is normally restricted to a very shallow layer at about 1.5 km (850 mb), assuming that transport takes place only above the trade-wind inversion. Above 3 km in altitude (700 mb), westerly winds ('counter trade-winds') prevail. Thus the transported mass of mineral dust is large during summer, as indicated by the seasonal variation of the turbidity coefficient B (for λ = 500 nm). B is a measure of the total aerosol mass in a vertical column for the particle-size range 0.1 µm $\leq$ r $\leq$1.0 µm and has been derived from a two-year series of measurements carried out at Sal by Jaenicke et al. (1977).

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[1]According to "Mean monthly climatic data for the world", 1965-1970, Asheville, N.C.

Until 1973, most of the physical and chemical data had been obtained from samples collected with nylon nets (Delany et al.,1967 and Parkin et al., 1970). Their collection efficiency, estimated by Parkin et al. (1970), was assumed to be 100% for particles with $r > 3$ µm. Below 3 µm radius, the collection efficiency was supposed to drop off, thus the submicron fraction is practically lost. Although this technique provides high flow rates, its collection efficiency as a function of particle size has never been investigated thoroughly. Thus large uncertainties have to be considered when referring to these data. Correction factors varying from 2 to 20 (compiled by Rahn et al., 1976) have to be applied to mass concentration data from nets. A comparison of the composition of high-volume filter samples taken near the West African coast by Lepple (1975) and the composition of samples taken by nets showed that the relative composition of the giant particles is fairly accurate. The absolute concentrations, however, are inaccurate.

This deficiency indicated a need for measurements of the physical and chemical characteristics of the NE trade-wind aerosol using refined sampling equipment in order to cover the whole particle-size range from Aitken nuclei ($r < 0.1$ µm) up to particles with $r > 100$ µm. Those measurements were performed at Sal in the Cape Verde Islands in 1973 (Jaenicke et al., 1977) to study the impact of Sahara dust on the maritime aerosol near its source. A second experiment was carried out on board the German research vessel 'Meteor' (cruise No. 32) crossing the North Atlantic from the Caribbean Sea to the West African coast at 15°N latitude. In this way the surface aerosol and its Saharan component (Schütz, 1977) were studied as a function of distance from the source.

The main results for the mineral component are as follows: Number-size distributions of total aerosol clearly showed no influence due to

dust-laden air masses for particles smaller than about 0.3 μm and larger than about 50 μm radius. This finding is best documented by mineral volume distributions (Figure 4), which demonstrate that the Sahara dust component is restricted to the size range $0.3\ \mu m \leq r \leq 50\ \mu m$. The concentrations of the submicron particles show relatively small variation over the entire NE trade-wind zone whereas the giant particles vary strongly. This behavior indicates different transport processes for each size fraction. Evidence for this hypothesis was observed during the following experiments. During the course of measurements in two cases a time shift in reaching a concentration maximum was observed when a dust cloud passed the sampling site. At the surface, particles with $r > 20\ \mu m$ reached their concentration peak 12 to 24 hours earlier than did the submicron particles. Apparently this time shift was due to different vertical transport processes, which supply the surface aerosol with mineral particles from the main dust reservoir above the inversion layer, as mentioned above earlier. These observations indicated a need for further investigations of the dust concentration versus altitude. To close this information gap and to understand better the main features of transport, a model was developed.

2). Transport Model

Models of Sahara dust transport have been developed by Fortak (1957) and Parkin (1974) with special emphasis on dustfall near the West African coast. Both models, however, appear to be insufficient to simulate dust transport properly. The model of Fortak (1957) considers both horizontal transport by wind and vertical turbulent diffusion and sedimentation of dust particles. Its disadvantage is the assumption that profiles of vertical wind and turbulent diffusion coefficients are constant

with altitude. Therefore, the specific horizontal flow conditions and the presence of the trade-wind inversion layer had been neglected. Furthermore, in Fortak's model a vertical dust concentration profile controlled by sedimentation-diffusion-equilibrium at the source was assumed. This, however, is in contrast to the strong vertical mixing of air observed over the Sahara by Carlson et al. (1972); this mixing requires that the vertical dust mixing ratio is constant with altitude. The model of Parkin (1974) considers vertical transport of particles by sedimentation and horizontal transport by wind only. Its application is therefore restricted to near-coastal transport of particles with $r > 10$ µm.

An improved Sahara dust transport model was developed by Schütz (1977). This model, a steady-state two-dimensional version, considers the same basic transport processes as did Fortak (1957) - i.e., easterly horizontal transport by zonal winds and vertical transport by turbulent diffusion and sedimentation of dust particles. In order to properly describe flow conditions in the NE trade-wind zone, however, actual vertical wind profiles up to 10 km altitude were used. The diffusion coefficient for vertical exchange was calculated from temperature and wind profiles. (Radiosonde data from Sal were assumed to be representative of the total transport distance over the Atlantic.) By this means inhibition of the vertical exchange between the trade-wind boundary and the Saharan air layer above by the trade-wind inversion layer could be incorporated into the model. The starting point of the model was fixed at the West African coast. There a dust layer was assumed to be between 1.5 km and 5 km in altitude (based on the picture presented in Carlson et al., 1972), with a constant vertical dust mixing ratio. The boundary layer (0 to 1.5 km) was assumed to be free of dust. This assumption was made on the basis that (1) air-mass trajectories never originate from the continent,

and (2) average surface winds close to the African shore are westerly. For the upper limit of the dust layer, 5 km altitude was selected as an average value, which is confirmed by Carlson et al. (1972) and by aircraft observations near the African coast. Other transport processes like subsidence and convection are assumed to balance each other. Removal of particles by precipitation is not considered, because the dust transport occurs primarily in dry continental air masses above the moist and cool boundary layer.

The model predicts the mixing ratio for the mineral particles as a function of the air-mass trajectory distance from the source area, altitude and particle size. Particle radii from 0.1 μm to 20 μm were considered. In order to study size distribution as a function of transport distance, an initial distribution at the coastline had to be assumed. According to measurements of the mineral aerosol size distributions in the Libyan desert (Schütz et al., 1974), an initial power function distribution[2] with an exponent of $\nu^* = 2$ seemed appropriate.

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[2]Approximations of most aerosol size distributions for particles with $r > 0.1$ μm can be obtained by a power function:

$$\frac{dN}{d\lg r} = n^*(r_o)\,(r/r_o)^{-\nu^*}$$

3). Results

a. Vertical mass distributions

For the vertical distribution of mineral dust within the Sahara plume, only a few measurements are available which allow a comparison between mass mixing ratios for mineral dust in the trade-wind boundary layer and in the Saharan air layer. Near the Canary Islands off the West African coast, size distribution measurements as a function of altitude have been performed by Gravenhorst (1975). He observed in the size range of 0.3 μm ≤ r ≤ 2 μm an increase of between one and two orders of magnitude in the mass mixing ratio as one went from surface air to elevations greater than 1.5 km. This strong difference could be related to dust- laden African air masses. Prospero et al. (1972) found during aircraft measurements near Barbados (5000 km from the Sahara) differences of only, at most, a factor of 4 between mass mixing ratios at the sea surface and in the Saharan air layer above. The large differences in the vertical mixing ratios near the source and the smaller differences after 5000 km of horizontal transport indicate a gradual downward mixing of dust from the plume. This idea is supported by the results of the transport model.

Figure 5 shows how according to the model the Sahara dust plume develops during transport and how the mass mixing ratio of the particles changes as a function of altitude and distance from the source. The mass mixing ratio is normalized to the initial mass mixing ratio. The submicron particles (0.1 μm ≤ r ≤ 1.0 μm)(which cause atmospheric turbidity) and those in the range of 0.1 μm ≤ r ≤ 20 μm - which contain most of the total mineral mass - behave differently. Up to a transport distance of 5000 km, the submicron particles are only inappreciably removed from the plume. The mass mixing ratio within the plume is reduced to only about 70% of the

initial value and at sea level the mass mixing ratio increases as the plume moves westward. These particles penetrate only very slowly by turbulent mixing and sedimentation from the elevated reservoir into the boundary layer. Therefore these particles are subject to long-range transport.

In contrast, the vertical distribution of total aerosol mass ($0.1\ \mu m \leq r \leq 20\ \mu m$) shows much more rapid variation with increasing transport distance. Due to the relatively large sedimentation velocities of giant particles, most of their mass falls out of the plume within the first 1000 km. A nearly constant vertical mass mixing ratio is achieved at a distance of approximately 1000 km. Thus the dustfall area is restricted to the first 1000 km of transport, which agrees with earlier observations by Semmelhack (1934).

b. Volume and mass concentrations in surface air

In the previous section a general survey of the development of the plume was given. Now we will discuss calculated volume and mass concentrations at sea level for different particle size ranges as a function of distance from the source. Figure 6 shows volume concentrations $V(r \geq 0.1\ \mu m)$, normalized to the maximum value of total volume concentration, for several particle size ranges at transport distances between 100 and 6000 km. The data plotted in Figure 6 are for power-function size distributions with exponents of $\nu^*=2$ and $\nu^*=3$ assumed for the initial distribution at the source.

Volume concentration ratios in the submicron particle range increase very slowly with distance from the source, especially for an initial distribution with $\nu^*=2$. Up to a transport distance of 6000 km, these particles make up only a few percent of the maximum volume concentration

(at 200 km). If an initial distribution with $\nu^*=3$ is assumed, however, considerably higher volume concentration ratios are reached with increasing distance from the Sahara. In this case the submicron particles may contribute more than 50% of the total volume at distances greater than 4000 km from the source. But it should be pointed out that in both cases the concentration ratios of the submicron particles are comparatively low within the first hundreds of kilometers of transport. As mentioned above, this indicates that these particles reach the surface layer only by slow vertical mixing.

The volume concentration ratios of particles with $r > 10$ µm increase up to 200 km from the source and thereafter decrease rapidly with distance. At 1000 km from the source their values are on the order of a few percent. At further distances their contribution to the total concentration is practically negligible. Independently of the assumed initial distribution, these particles, which sediment quickly, are deposited within 1500 km of the Sahara.

Particles in the intermediate size range ($1\ \mu m \leq r \leq 10\ \mu m$) increase in relative values up to 1500 km, where a maximum of approximately 50% of the total volume is reached. For distances larger than 1000 km their concentration decreases but still accounts for most of the mineral volume which is transported over larger distances. In the case of calculations with $\nu^*=2$, 90% of the total volume transported over distances larger than 1500 km consists of these particles. For $\nu^*=3$ the contribution of this particle range is smaller because of rapidly increasing volume fractions of submicron particles at distances greater than 2000 km from the Sahara.

To verify the model calculations, a comparison with actual mass concentrations measured at sea level over the Atlantic Ocean will be given. Data were collected by the authors during field studies of the

NE trade-wind aerosol over the Atlantic as mentioned above. Figure 7 shows measured and calculated concentrations for the submicron and giant particles as a function of distance from the source. As above, initial distributions with $\nu^*=2$ and $\nu^*=3$ will be distinguished. Calculated mass concentrations are normalized to M ($0.1\ \mu m \leq r \leq 20\ \mu m$) = $100\ \mu g\ m^{-3}$ at a distance of 1000 km from the source. This value was selected from mean mass concentrations of our Atlantic data. From Figure 7 some important conclusions can be drawn about the shape of the assumed initial size distribution at the source. Measured mass concentrations agree reasonably well with calculationsusing $\nu^*=3$. For the submicron range, however, the model with $\nu^*=3$ predicts concentrations approximately one order of magnitude higher than observed. A better assumption for the initial distribution within the size range of $0.1\ \mu m \leq r \leq 20\ \mu m$ would be $\nu^*=2$. In that case the measured mass concentrations for both size ranges are predicted better by the model, as seen in Figure 7. The assumption of $\nu^* = 2$ at the source is confirmed by measurements of mean mineral size distributions for $r \geq 1\ \mu m$ over the Libyan Sahara by Schütz et al. (1974). A similar value of $\nu^*=2$ has been obtained by Gillette et al. (1972) for soil-derived aerosol in Texas and Nebraska. Thus, this value may be typical of aerosol generated from soil in many regions of the globe. Figure 7 shows that neither the total sea level mass nor its submicron fraction decrease strongly beyond 1000 km from the source. In contrast, significant variations in the mass concentration have to be expected within the first thousand kilometers of transport. Unfortunately, no data are avilable to check this idea. The rapid drop-off of mass concentrations of the submicron fractions observed beyond 3000 km is due to a short-term interruption of the steady Sahara air outflow by strong low pressure systems.

c. Volume distributions in surface air

One goal of this investigation was to study the development of the size distribution at sea level as a function of distance from the Sahara. For a better understanding of the main features of the measured size distributions across the Atlantic, a comparison with modeled distributions will be discussed. The calculations are based on an initial distribution at the source with $\nu^*=2$ according to the results displayed in Figure 7. The size distributions were then converted to volume distributions so that important variations in the giant particle range could be more closely observed.

Figure 8 shows a comparison between measured and calculated volume distributions for selected distances from the source. From this figure it appears that the assumed initial distribution at the source is a reasonably accurate representation of the actual distribution observed at the Sahara Desert. For movement westward from the source, the model indicates a shift of the maximum of the distributions toward smaller particle radii, with a general decrease in the maximum concentration. These results agree qualitatively with the measured size distributions. According to these data, the maximum of the volume distribution at the source is at 50 μm radius and subsequently shifts to radii of approximately 10 μm, 4 μm and 1 μm for transport distances of 1000 km, 2000 km and 5000 km, respectively. The volume distribution observed for the mineral aerosol over the Caribbean Sea, 5000 km from the Sahara Desert, may represent background conditions for this aerosol. Finally, it has to be pointed out, however, that the model is not able to explain the comparatively high concentrations of particles of radius larger than 10 μm observed during actual field measurements. These giant particles may be

agglomerates of smaller particles formed during coalescence of cloud droplets and raindrops, which later evaporate to dry residues. This idea has been supported by microscopic investigations (by the authors) of those particle fractions which consist entirely of agglomerates.

d. Mineral mass transport budget

The transport model offers the possibility of estimating the output of mineral particles for the Sahara Desert. This can be done on the basis of a series of turbidity measurements from 1973-1975 (Jaenicke et al., 1977) at Sal which allows relative values of dust released from the source as predicted by the model to be transformed into absolute masses as a function of transport distance. Once the mass in a vertical column is fixed, the fractions of the total mass which are deposited into the ocean as well as those remaining airborne can be determined as a function of the distance from the Sahara.

According to the turbidity measurements of Jaenicke et al. (1977), a mean vertical columnar particle mass load $M_c(0.1\ \mu m \leq r \leq 1.0\ \mu m) = 9.1 \times 10^{-6}\ g\ cm^{-2}$ could be estimated for Sal, at about 1000 km from the source. This columnar mass consists practically only of mineral particles. Other components, like sea-salt, $(NH_4)_2SO_4$ and organic aerosol fractions, are only minor constituents of the surface aerosol and do not contribute very much to the total mass load of the column of 5 km altitude (Jaenicke et al., 1977). It was shown in Section (a), that particles in the size range $0.1\ \mu m \leq r \leq 1.0\ \mu m$ are essentially not removed from the plume during the first 1000 km of transport. This allows one to assume the same figure for the source column between 1.5 km and 5.0 km altitude as above.

In Section (b) it was shown that the source aerosol can be approximated

best by a power distribution with $\nu^*=2$. In such an aerosol only 6.7% of the total mass is contained in the submicron range. This leads to a columnar mass of 1.35×10^{-4} g cm^{-2} for the entire size range of 0.1 μm ≤ r ≤ 20 μm of the source aerosol. On the basis of a transport channel of 1000 km width (15° to 24°N) and 3.5 km height (1.5 km to 5.0 km altitude) and assuming an average vertical zonal easterly wind of 6 m s^{-1} (Newell et al., 1972), the amount of mineral dust transported per year over the North Atlantic can be estimated.

This amount is given in Figure 9, which is the mass budget of the Sahara dust plume. Roughly 260×10^6 tons yr^{-1} of mineral dust leave the source. Within about 300 km of the West African coast westerly winds dominate the near-surface flow, as mentioned above; this return flow probably carried about 90×10^6 tons yr^{-1} of dust back toward the desert. Lepple (1975) estimates an annual dust output at the West African coast between 15° and 30° N of roughly 480×10^6 tons. This figure is based on mass concentration measurements near Cape Blanc, mean zonal wind velocity, and a dust layer height of 1 km, and is equivalent to 32×10^6 tons yr^{-1} for one degree of latitude. For a 1000-km - wide transport channel, this figure leads to about 290×10^6 tons yr^{-1}, close to our estimate.

A net 170×10^6 tons yr^{-1} remains airborne and is transported to the west. Because of the rapid loss of giant particles by sedimentation, mentioned in Sections (a) and (b), approximately 2/3 of the total Sahara aerosol mass falls out within the first 1000 km from the coast. At 1000 km only about 80×10^6 tons yr^{-1} remain airborne. The budget predicts only comparatively small variations for subsequent westward motion.

Beyond 1000 km from the Sahara, the aerosol consits mainly of submicron particles which are only slowly removed from the plume by vertical mixing. According to the model roughly 50 x 10^6 tons yr^{-1} of mineral dust from the Sahara Desert will be transported over the Caribbean Sea. Prospero et al. (1972) found a semi-annual flux of roughly 37 x 10^6 tons. These two figures for the Caribbean Sea are in reasonable agreement, especially when the general uncertainty of these estimates is considered.

Prospero et. al. (1972) claim further that approximately the same quantity of dust is deposited in the Atlantic between western Africa and the Caribbean. This, however, seems unlikely according to our model, which predicts that roughly 70% of mass emitted from the Sahara is deposited into the Atlantic and 30% flows beyond the Caribbean Sea. The main difference between the estimates of Prospero et al. (1972) and our model is our calculated rapid loss of mass during the first 1000 km of transport.

On the basis of the estimates by Prospero et al. (1972), Lepple (1975) and us (in this paper), a range of roughly 60 - 200 x 10^6 tons yr^{-1} can be calculated as the Sahara dust output to the North Atlantic. Compared to the estimated global production of mineral dust of 100-500 x 10^6 tons yr^{-1} (SMIC, 1971), our Sahara estimate indicates that the global flux figure maybe too low. Recently Junge (1977) calculated the mineral dust burden from global aerosol data and, assuming a mean residence time of one week, calculated a total tropospheric mineral dust production rate of 130 - 800 x 10^6 tons yr^{-1}. Using either global figure, however, it is clear that the Sahara Desert contributes significantly to the atmospheric mineral dust load.

e. Atlantic deep-sea sediments

The importance of airborne dust to the formation of deep-sea sediments on a global scale has been stressed by Griffin et al. (1968).

In particular for the NE trade-wind zone of the Atlantic, a contribution by eolian dust from the Sahara can be seen from the kaolinite pattern of the sediments. Considerable amounts of noncombustable (at 500°C) suspended matter in near-coastal waters of the Atlantic are related to Saharan mineral particles (Emery et al., 1974). Rona (1971) suggests that airborne African dust has, to a certain extent, formed the bottom morphology of the Cape-Verde Plateau, the shape of which is similar to the atmospheric dust-fall area and is favored by surface winds as well as ocean currents.

Particle mass distributions of sediment samples near the West African coast have been investigated by Beltagy et al. (1972). For comparison with the mineral aerosol their data of total particulate matter (on a carbonate-free basis) and of quartz have been converted into a power number size distribution. Within the size range of 1 μm ≤ r ≤ 10 μm and at a distance of 2500 km from the coast. the distribution of the sediments roughly corresponds to that of the mineral aerosol with a slope of $\nu^*=3$ (unpublished data of the authors). Size distributions of quartz particles from deep-sea cores of the Cape Verde Islands region by Dauphin (1977) also reflect the coarseness of the wind-transported material to the Sahara region.

The Sahara mass budget over the Atlantic (Figure 9) predicts a definite contribution to the deep-sea sediments by dry fallout from the Saharan plume. The mass deposited in the ocean can easily be converted into accumulation rates for the sediments as a function of transport distance. After assuming an appropriate value for the packing density of the sediment deposits, the accumulation rates can be expressed as cm/1000 yr. In this way our transport model can be used to predict details of sedimentation in the Atlantic. Calculated accumulation rates

for a packing density of 2 g cm^{-3} are plotted in Figure 10. According to these data, a zone of relatively high accumulation rates should be expected within the first 2000 km from the Sahara Desert. When most of the mass of the plume has fallen out (distances larger than 2000 km), a zone of comparatively low accumulation rates follows.

The calculations can be compared with actual sediment data representative of that area of the Atlantic, as compiled by Ku et al. (1968) (derived from Goldberg et al., 1963). The model overestimates the first 2000 km, whereas beyond 2000 km the accumulation rates are predicted reasonably. To this extent the model and the measured data are not in opposition. One value from the Cape Verde archipelago derived from Rothe (1973) indicates that our model predicts the accumulation at about 800 km from the source to within an order of magnitude. In this area, however, sedimentation near the continent and the islands might be influenced by a contribution of other non-airborne particulate matter, as pointed out by Rothe (1973). This has to be considered for values derived from Ericson et al. (1961), which refer to cores very near the coast. Their values near the continental rise range from about 2 to 45 cm 1000 yr^{-1} and might be affected by the ocean floor topography.

Additionally, an estimate based on atmospheric dry dust fallout for the near-coastal region by Lepple (1975) is given. This estimate is based on a suite of high-volume filter samples in surface air and assumptions about the vertical dust and wind profile, which introduce an uncertainty of at least a factor of two. According to this calculation, a strong decrease in the accumulation rates near the West African coast should be expected. Due to the lack of actual data within this area, the validity of these rough estimates has not yet been proven and can be taken as a first guess only. Furthermore, it should be noted that our model is

based on mean recent atmospheric flow conditions. The literature values used for comparison reflect a much longer time scale. Finally, one has to keep in mind that when the airborne particles precipitate as deep-sea sediments, currents and other processes in the ocean, might alter their physical characteristics.

Conclusions

The transport model presented here offered the possibility of studying the development of the dust plume as a function of distance from the source. It shows the different behavior of the various particle size ranges as they travelled over the Atlantic. Submicron particles are predicted to remain in the dust plume for long distances. Their removal, which is caused mainly by turbulent diffusion, is hindered by the trade-wind inversion layer, which suppresses vertical exchange and leads to a delayed downward flux of this size range. In contrast, the giant particles sediment rapidly and are unaffected by the inversion layer; they are removed from the plume in relatively short distances. Within 1500 km from the Sahara these particles, which contain most of the mineral aerosol mass, have fallen out. Comparison between measured and calculated mass concentrations showed that reasonable agreement can be achieved by assuming an initial size distribution with $\nu^*=2$. This indicates that giant particles are abundantly emmitted by the source, which confirms earlier findings by the authors.

The budget calculation for mineral mass transported over the Atlantic indicates that previous mineral source-strength estimates both for the Sahara Desert and for the world as a whole must be revised upward. Because the Sahara Desert is only one of many sources of mineral dust, and because practically nothing is known about the dust output of other

deserts, we are still far away from a reliable estimate of global source strength for mineral aerosols.

Finally, it has to be pointed out that this model must be taken as a preliminary approximation reflecting rough average atmospheric conditions. For better transport predictions by such a model, however, a need for thorough investigations of the source itself is apparent. There is a great lack of knowledge about the vertical distribution of mineral dust as well as its size distribution. Furthermore, the dust generation conditions have not yet been investigated for different soil types and regions in the Sahara. These studies would contribute to finding more realistic boundary conditions.

To study long-term dust output variations, size distribution measurements on deep-sea sediments for particles with $r > 0.1$ μm, which are not altered by physical and chemical processes in the ocean, could be very useful.

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Captions

Fig. 1: Historical map of dustfall records around the world (compiled by Ehrenberg, 1862). Dotted areas indicate dustfall regions. Near the West African coast an extensive area can be seen where dust originally from the Sahara Desert was very frequently observed falling out on cruising ships.

Fig. 2 Isolines of Linke's turbidity factor taken as a first guess for the source region of dust transported over the ocean. Turbidity factors calculated from sunshine recorder data (using values from Dubief, 1959) and direct measurements by Dubief (1959). For details, see Jaenicke (1977).

Fig. 3 Comparison between mean annual frequency of sandstorms according to Dubief (1953) for different regions of the Sahara Desert, vertical wind field over the Atlantic and atmospheric turbidity derived from measurements by Jaenicke et al. (1977). A possible explanation for the annual variation of turbidity, which is a measure of the atmospheric dust load, is the superposition of sandstorm frequency at the source area and of the vertical wind field over the ocean.

Fig. 4 Mineral aerosol volume distributions in surface air for various distances from the Sahara. Measured data by Jaenicke et al. (1977) and Schütz (1977).

Fig. 5 Vertical distributions of mineral dust for selected transport distances from the source. Vertical mixing ratios are plotted for the mass of mineral dust particles in the submicron range (0.1 μm ≤ r ≤ 1.0 μm) and for the total mass (0.1 μm ≤ r ≤ 1.0 μm) normalized to the initial mixing ratio at the source (x = 0 km). The vertical distributions of the submicron particles remain nearly unchanged over large transport distances, whereas those for the total mass show significant modifications up to approximately 1000 km distance. Thus most of the total mass is removed from the plume near the source but submicron particles are subject to long range transport.

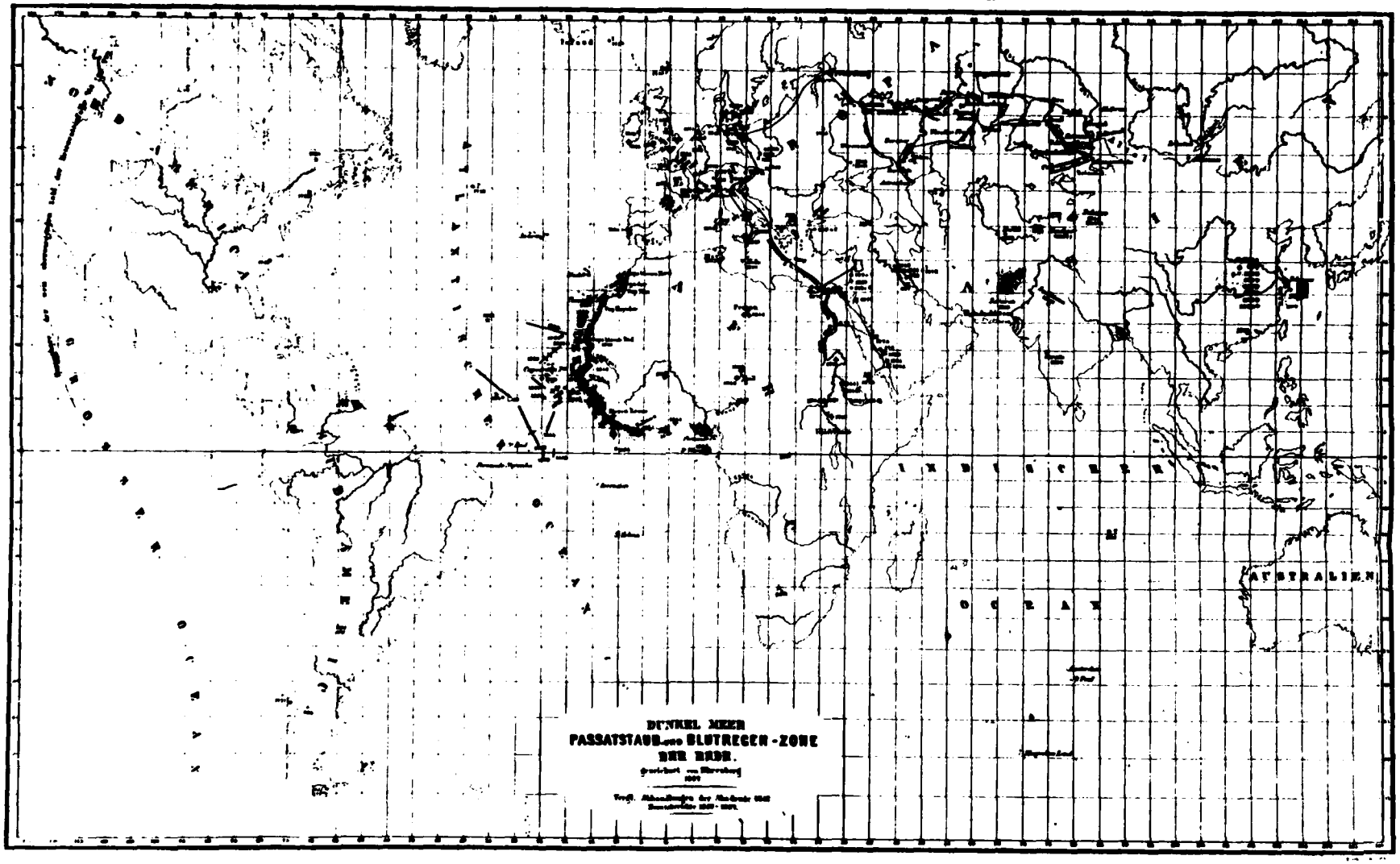
Fig. 6 Volume concentration ratios at sea level for different particle size ranges as a function of the distance from the source. Model calculations for assumed initial size distributions with $\nu^* = 2$ and $\nu^* = 3$. Except for particles with $r \geq 10$ μm the graphs indicate different volume concentration ratios for the size ranges considered when moving westward under the plume.

Fig. 7 Mineral mass concentrations at sea level as a function of the transport distance from the Sahara. Comparison between calculated data (normalized to M (0.1 μm ≤ r ≤ 20 μm) = 100 $\mu g/m^3$ at a distance of 1000 km) and data measured by the authors for the submicron and the total mass range. The measurements can best be described by the calculations when assuming an initial distribution at the source with $\nu^* = 2$.

Fig. 8 Mineral volume distributions at sea level for various distances from the Sahara. The dotted lines represent calculations assuming an initial distribution with $\nu^* = 2$ at the source. The heavy lines represent measurements of mean volume distributions at the source and for selected transport distances over the Atlantic. Both measurements and calculations indicate a shift of the maximum of the distributions toward smaller particle radii.

Fig. 9 Annual mass budget of dust transported over the Atlantic in the NE-trade wind zone (model).
Most of the mineral mass emitted by the desert will be deposited in the ocean within the first 1000 km of transport. For larger distances only slight variations of the budget can be expected.

Fig. 10 Accumulation rates of deep-sea sediments of the Atlantic due to Saharan dust input.
Despite the deviations between measured and calculated data, the model predicts the sedimentary deposition of the dust fairly realistically.


AUSTRALIEN
O C E A N
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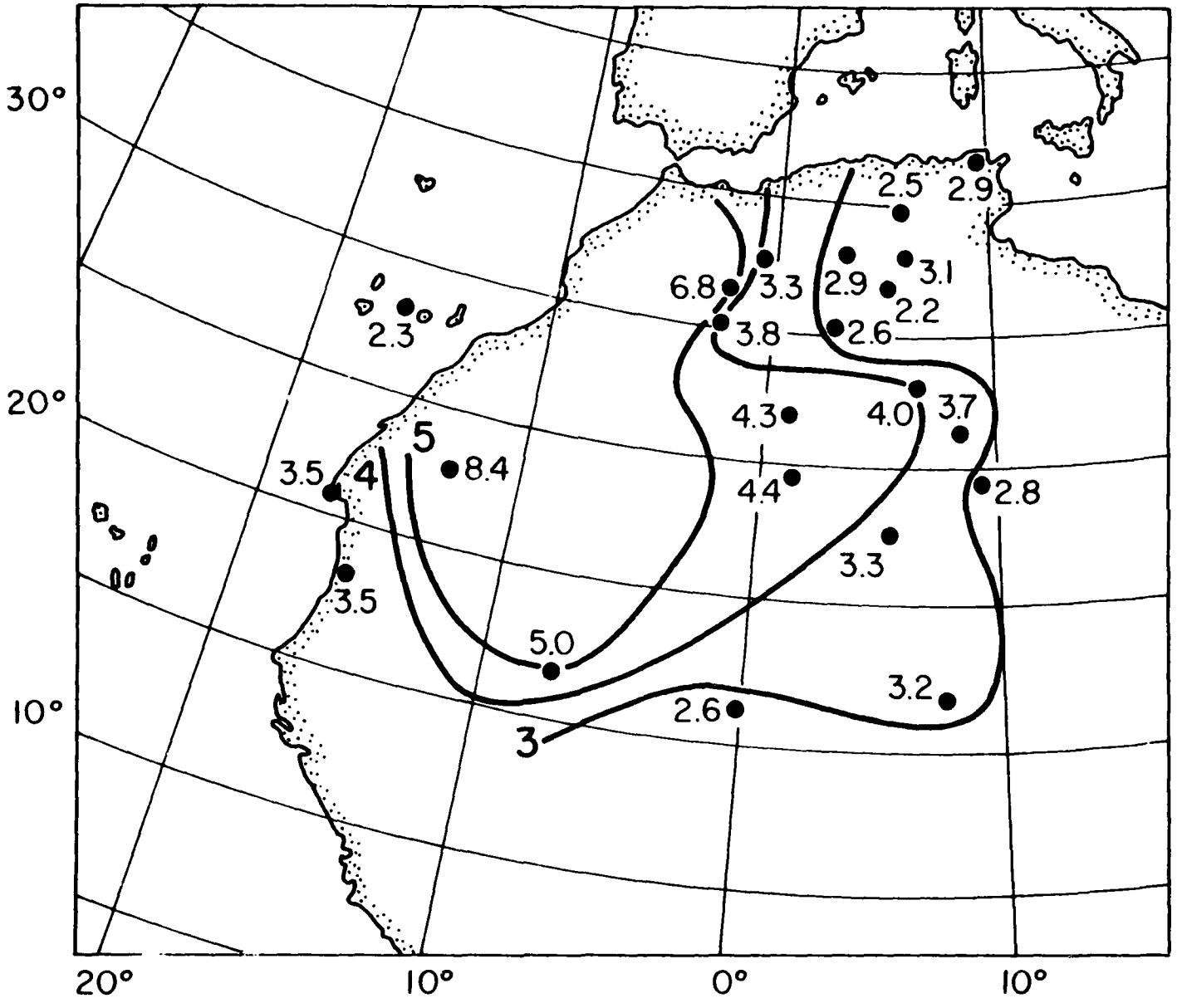
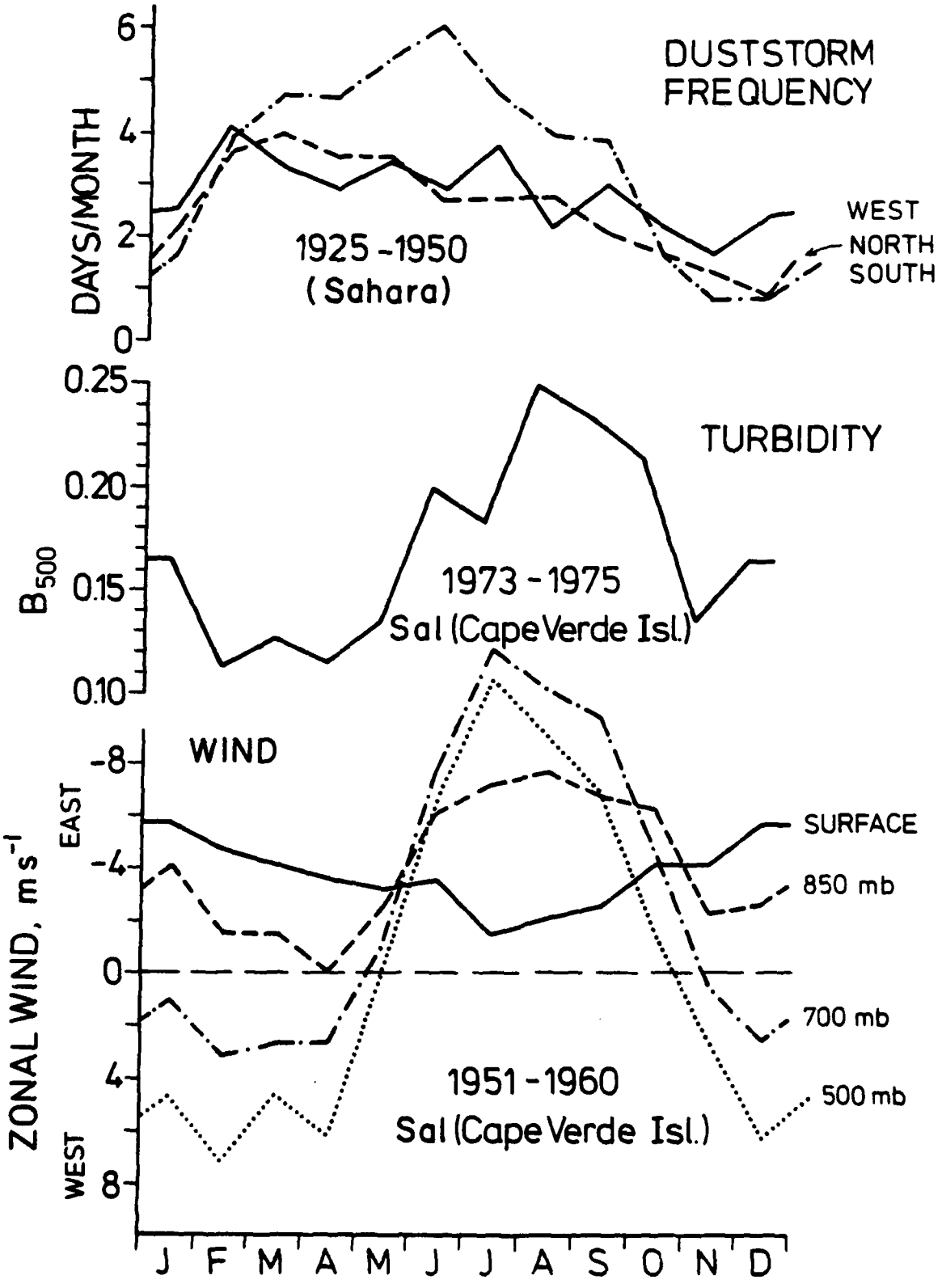
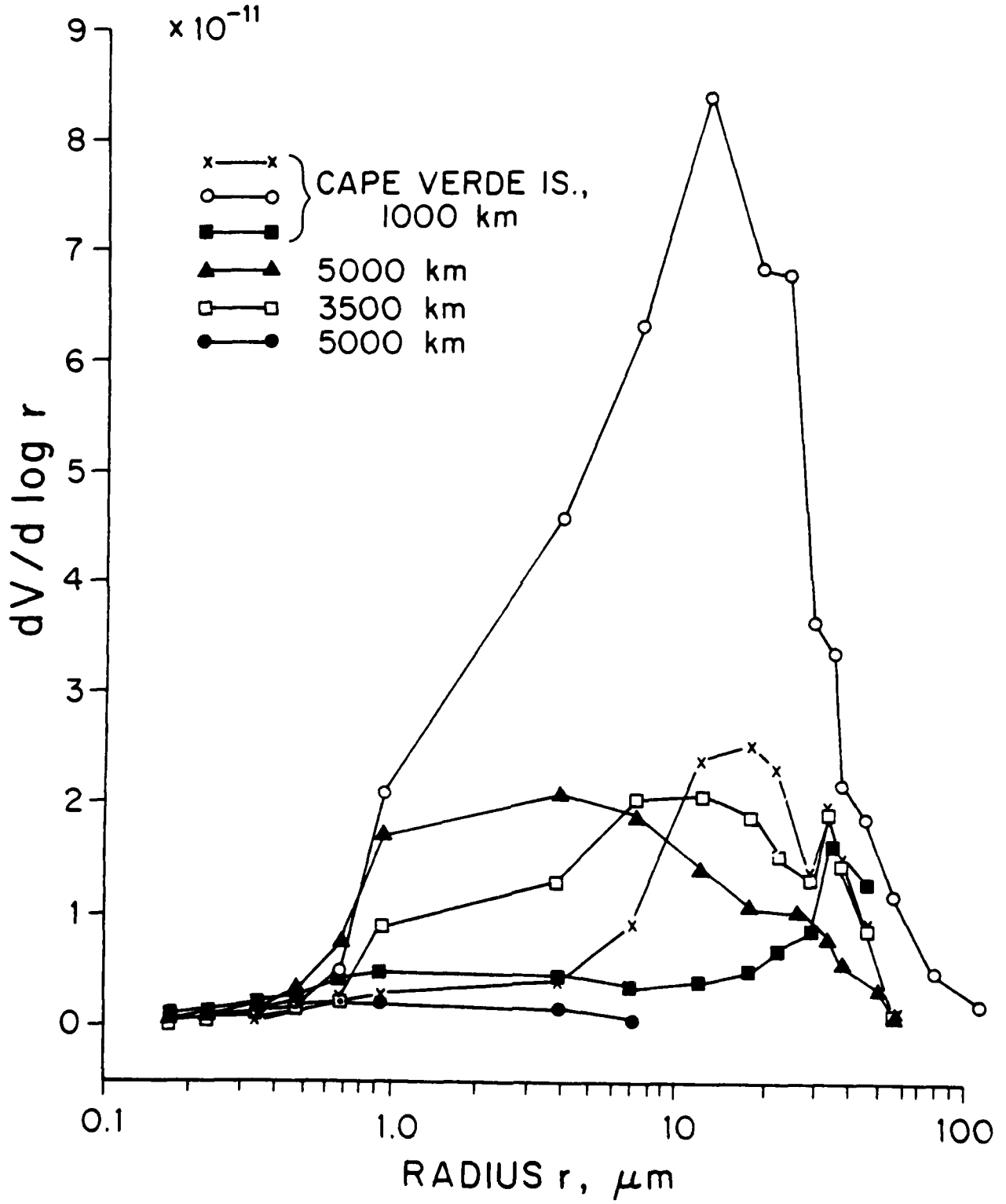
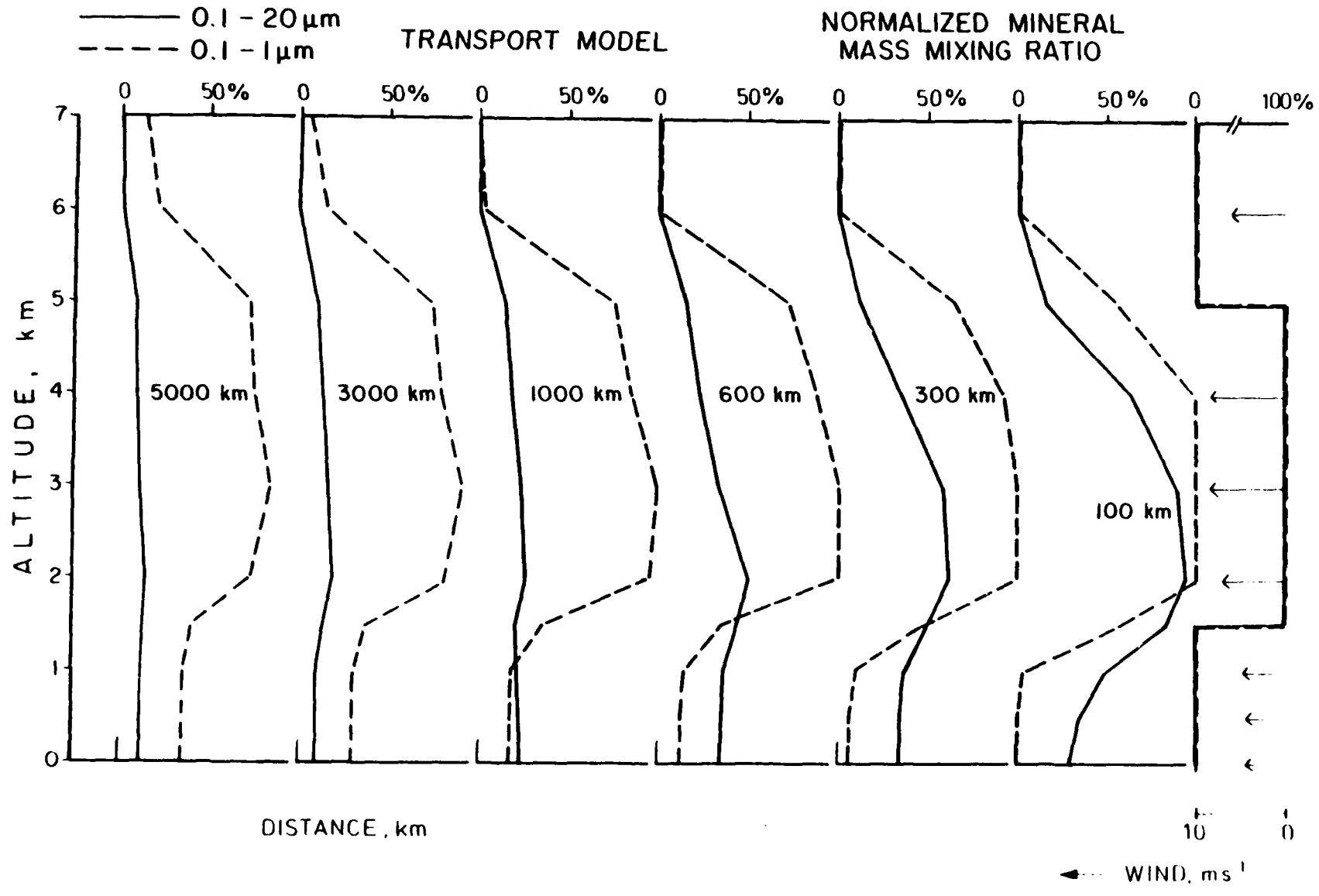
PASSATSTAUB... BLUTREGEN-ZONE
DER ERDE.

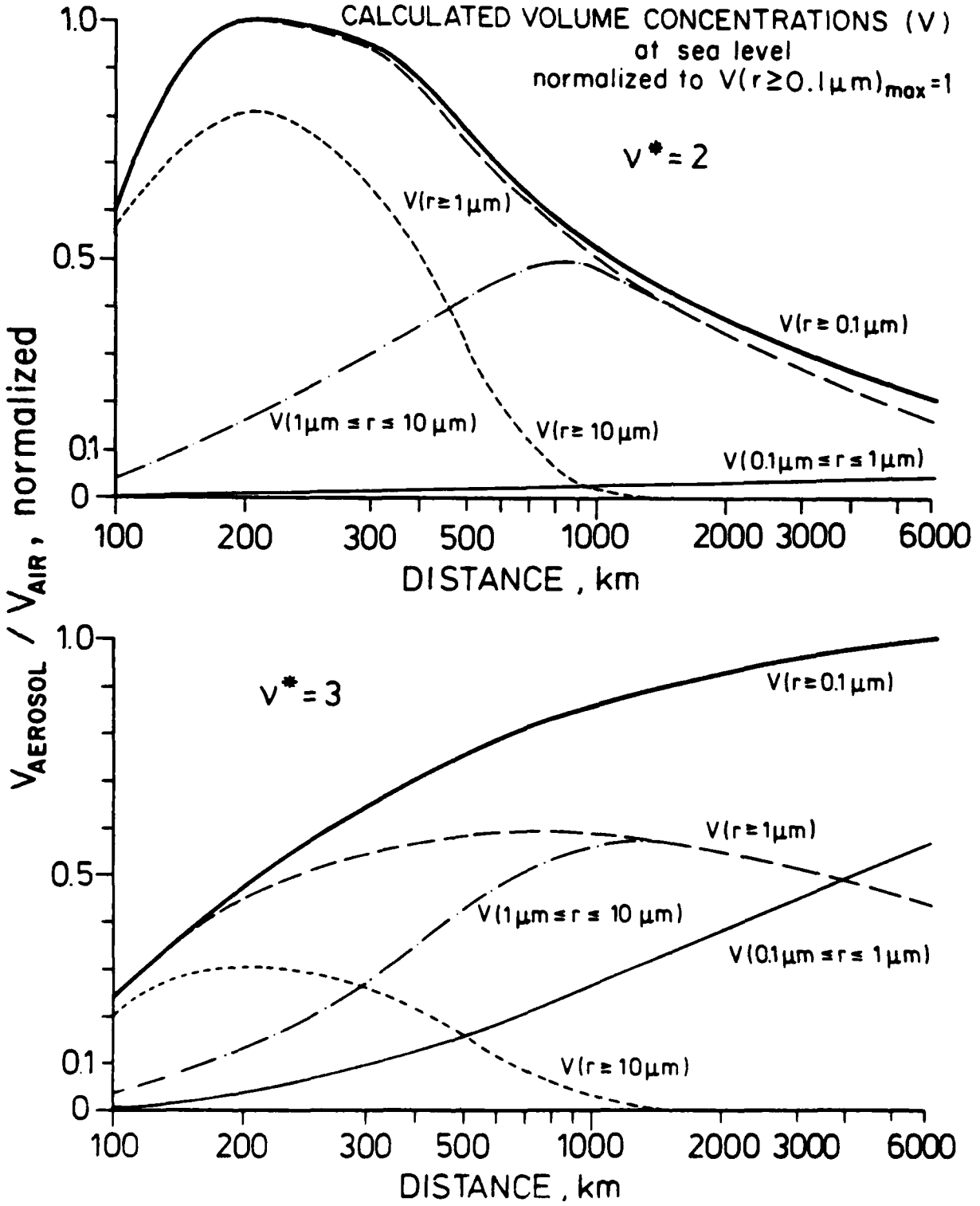
ATMOSPHERIC TURBIDITY

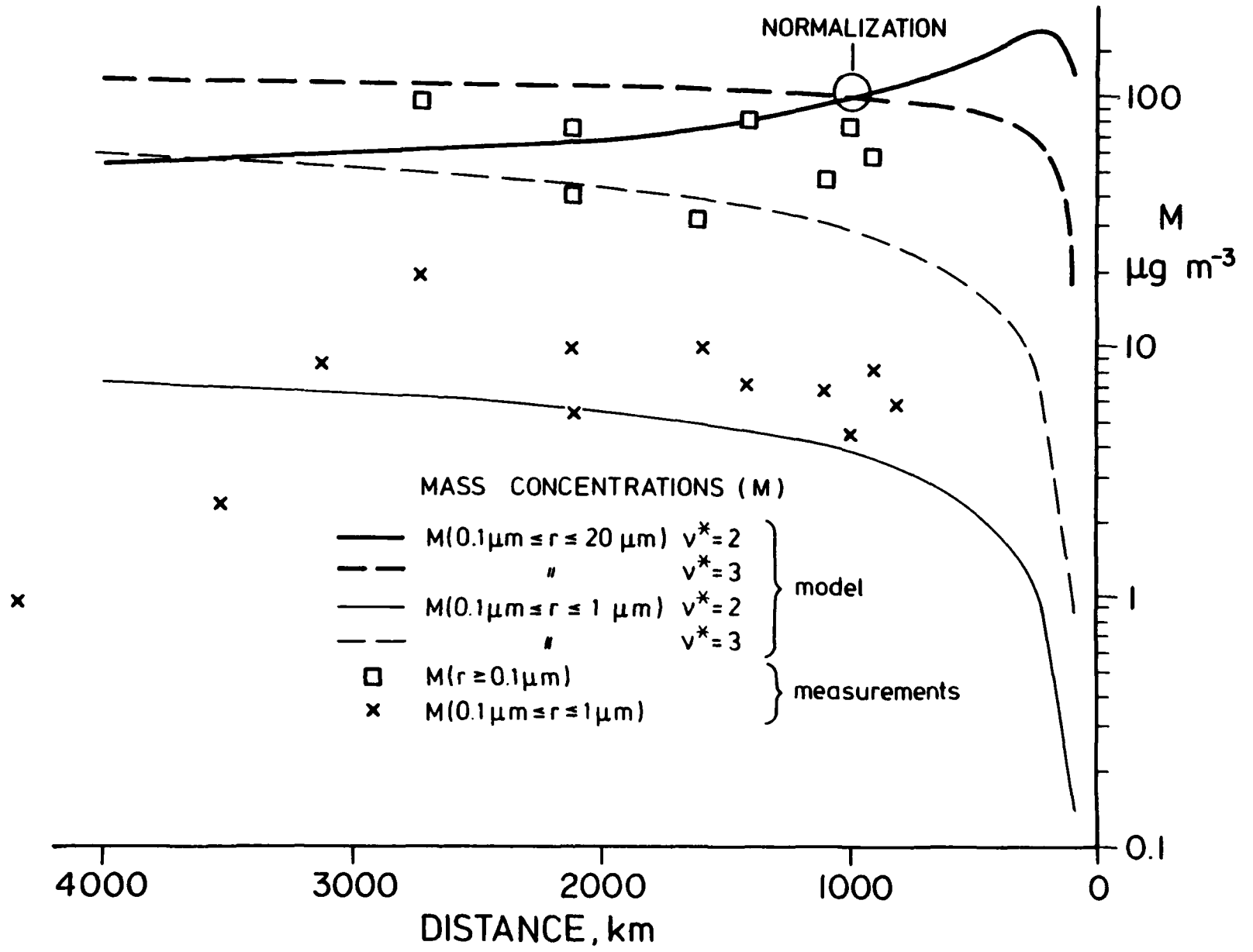


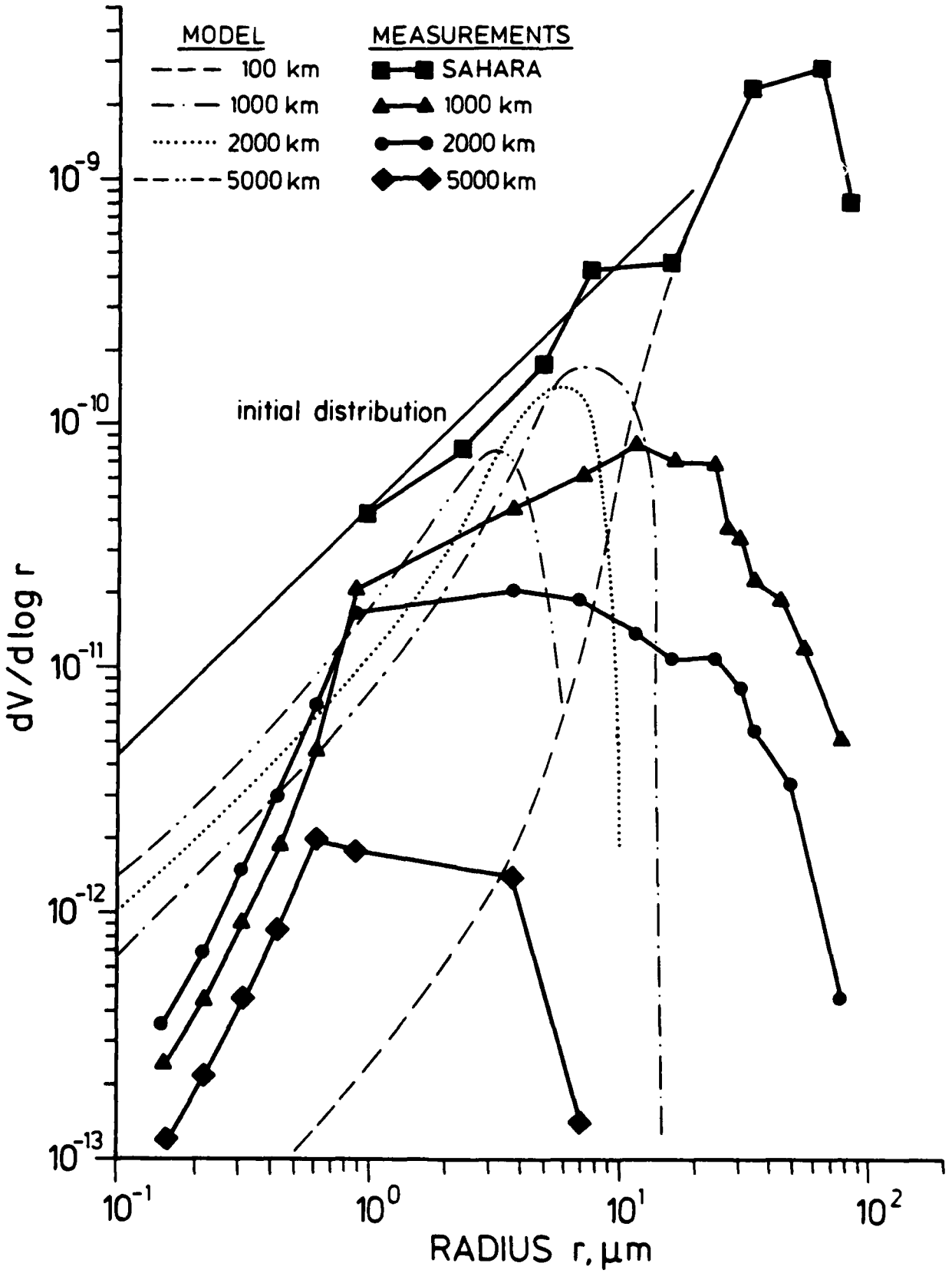

DUSTSTORM
FREQUENCY
DAYS/MONTH
6
4
2
0
1925 -1950
(Sahara)
WEST
NORTH
SOUTH
TURBIDITY
B_{500}
0.25
0.20
0.15
0.10
1973 - 1975
Sal (Cape Verde Isl.)
WIND
ZONAL WIND, $m\,s^{-1}$
EAST
WEST
-8
-4
0
4
8
SURFACE
850 mb
700 mb
500 mb
1951 - 1960
Sal (Cape Verde Isl.)
J F M A M J J A S O N D


x 10⁻¹¹
9
8
7
6
5
4
3
2
1
0
dV/d log r
CAPE VERDE IS., 1000 km
5000 km
3500 km
5000 km
0.1
1.0
10
100
RADIUS r, μm


0.1 - 20 μm
0.1 - 1 μm
TRANSPORT MODEL
NORMALIZED MINERAL
MASS MIXING RATIO
0
50%
100%
ALTITUDE, km
0
1
2
3
4
5
6
7
5000 km
3000 km
1000 km
600 km
300 km
100 km
DISTANCE, km
10
0
WIND, ms⁻¹


CALCULATED VOLUME CONCENTRATIONS (V)
at sea level
normalized to V(r≥0.1μm)max=1
ν*=2
V(r≥1μm)
V(r≥0.1μm)
V(1μm≤r≤10μm)
V(r≥10μm)
V(0.1μm≤r≤1μm)
1.0
0.5
0.1
0
100
200
300
500
1000
2000
3000
6000
DISTANCE, km
VAEROSOL / VAIR, normalized
ν*=3
V(r≥0.1μm)
V(r≥1μm)
V(1μm≤r≤10μm)
V(0.1μm≤r≤1μm)
V(r≥10μm)
1.0
0.5
0.1
0
100
200
300
500
1000
2000
3000
6000
DISTANCE, km

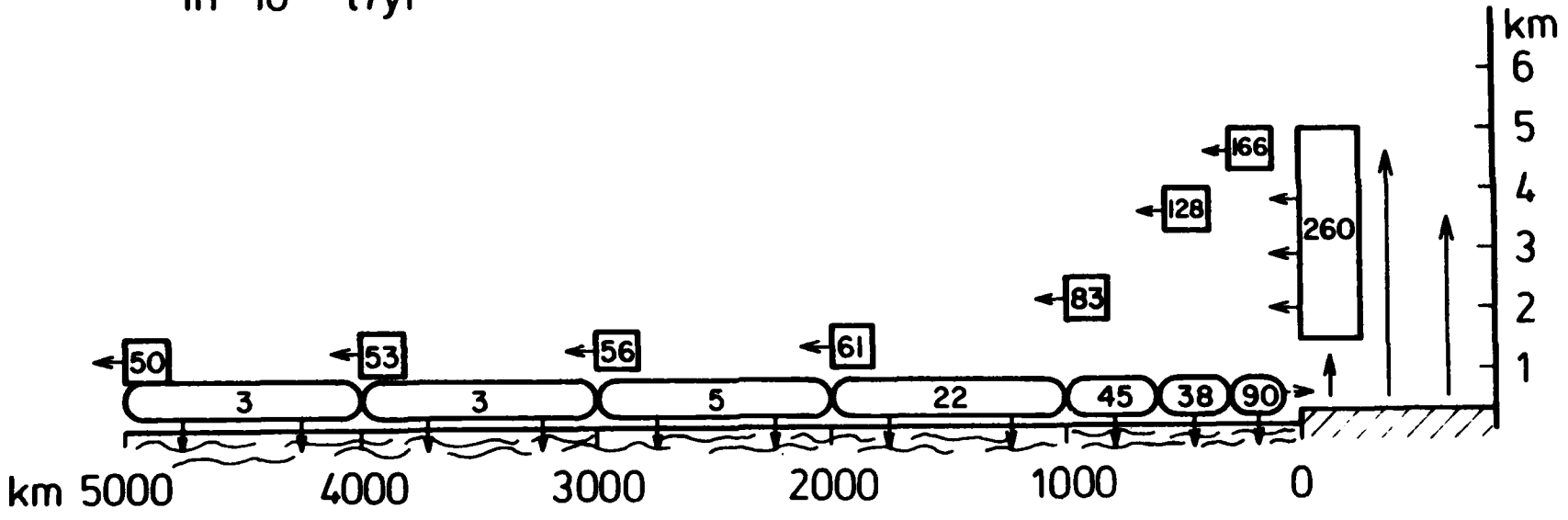

NORMALIZATION
100
10
1
0.1
M
μg m⁻³
MASS CONCENTRATIONS (M)
M(0.1μm ≤ r ≤ 20 μm) ν*=2
" ν*=3
M(0.1μm ≤ r ≤ 1 μm) ν*=2
" ν*=3
model
M(r ≥ 0.1μm)
M(0.1μm ≤ r ≤ 1μm)
measurements
4000
3000
2000
1000
0
DISTANCE, km


MODEL
100 km
1000 km
2000 km
5000 km
MEASUREMENTS
SAHARA
1000 km
2000 km
5000 km
initial distribution
dV/d log r
10^{-9}
10^{-10}
10^{-11}
10^{-12}
10^{-13}
10^{-1}
10^{0}
10^{1}
10^{2}
RADIUS r, µm

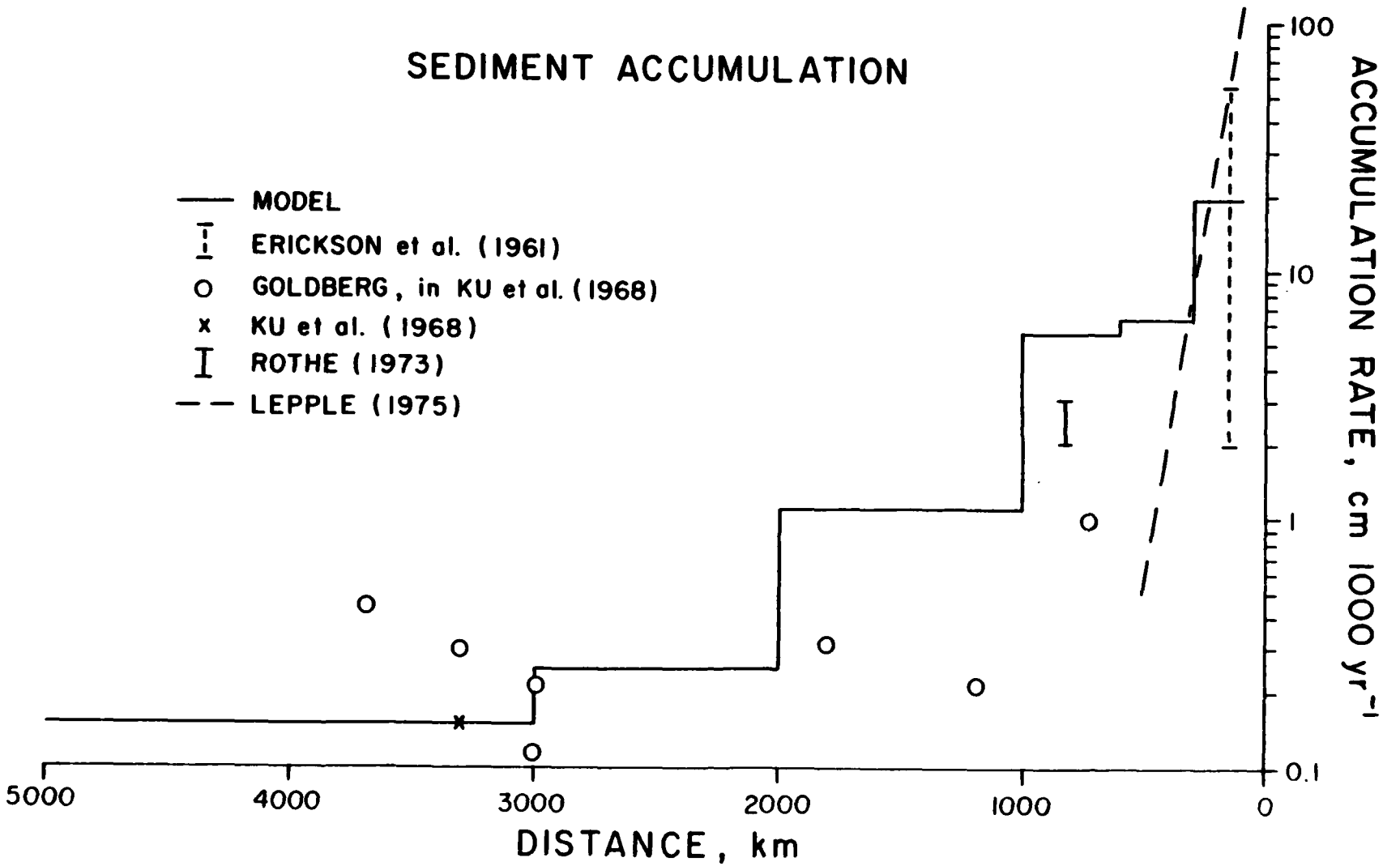
TRANSPORT OF SAHARA MINERAL DUST

15°N - 24°N ~ 1000km

in 10^6 t/yr



NORTH ATLANTIC OCEAN


SEDIMENT ACCUMULATION
MODEL
ERICKSON et al. (1961)
GOLDBERG, in KU et al. (1968)
KU et al. (1968)
ROTHE (1973)
LEPPLE (1975)
100
10
1
0.1
ACCUMULATION RATE, cm 1000 yr⁻¹
5000
4000
3000
2000
1000
0
DISTANCE, km

G. The improved (C117D) aircraft air-sampling system

Our first field experiment in Barrow took place in April and May 1976. The aircraft air-sampling phase of this experiment was done with a Cessna 180 into which a relatively small sampling system had been installed (Details are given in Section ID.). Briefly, it consisted of four air probes built into one of the side windows - small probes for a manual condensation-nucleus counter, a Nuclepore filter, and a Millipore filter, and a large probe for a high-volume cellulose filter. Air was drawn through the Nuclepore and Millipore filters by the aircraft's vacuum system, but the high-volume filter needed a vacuum pump which was powered by an inverter and batteries.

This system worked outstandingly well, and provided all the information upon which our ideas about Asian dust were based. It did, however, have some limitations which we feel should be overcome for subsequent experiments. For example, actual sampling times per flight were short - just over three hours. During periods when aerosol concentrations were low, we were forced to combine samples from two consecutive flights in order to get enough mass for analysis. Also, on at least one of these flights there seemed to be an aerosol layer above 3 km, the effective ceiling of the aircraft. Lastly, we wanted to take more than one high-volume sample at a time, for example to add a glass-fiber filter for determination of total aerosol mass.

In short, we wanted to be able to fly higher, longer, and to take more samples. When one of NARL's C117D's was made available to us, we decided to outfit it with an expanded and improved sampling system. This system was built mostly at URI and installed on the aircraft in July 1977, after a series of delays from the original proposed data of March 1977. In its final version it consists of 7 probes through the top of the fuselage just behind the navigator's cabin. There is a very small probe for an automatic

condensation-nucleus counter (Environment/One Model Rich 100 with recorder), two small probes for 47-mm Millipore and Nuclepore filters, and four large probes for 10-cm high-volume filter and impactor samples. As originally foreseen, there would be two filters (glass-fiber and cellulose) and two high-volume cascade impactors (with glass-fiber and cellulose impaction surfaces, respectively). After the system had been built, however, we discovered that the Sierra impactors which we had been using in other locations and were planning to use here seriously malfunctioned in dry atmospheres such as are found aloft in the Arctic. For this reason we have not yet used impactors on the system.

Air is drawn through the small Nuclepore and Millipore filters by 28V vacuum pumps powered directly by the aircraft's electrical system. Vacuum pumps for the high-volume samplers are also powered from the aircraft's electrical system, but through inverters. The complete system requires 5 1000-Watt inverters: one each for the four large probes and one for the condensation-nucleus counter and radiation instruments (to be discussed below). At the moment we have two inverters, are requesting funds for a third, and hope that an independent proposal to NSF will provide the remaining two.

The last part of the aircraft system is Dr. Shaw's hemispheric flux detector. This compact sensor has also been mounted on top of the fuselage, through the aft porthole. Its purpose is to obtain vertical profiles of integrated radiation fluxes through layers of Arctic haze, and from these data to derive a more accurate assessment of the perturbation of these layers on the radiation budget than is possible with sun photometer data alone.

In the future we hope to add a number of miscellaneous components to the system, such as a temperature-dewpoint sensor and more recorders.

H. July 1977 field experiment in Barrow

This experiment was originally intended for March 1977, to confirm the presence of haze layers of Asian origin in the Barrow atmosphere during spring, and possibly also to follow a haze layer southward from Alaska out over the Pacific, thus positively excluding Alaska as a source for the haze. A series of unavoidable scheduling problems with the C117D aircraft forced a postponement until July 1977, and also changed the character of the entire experiment. According to the ideas we had been developing during the first 18 months of this contract, the Barrow atmosphere in high summer should be much cleaner than in spring, with little or no Asian desert dust and greatly reduced levels of pollution aerosol present. We saw that a July experiment could check this. Further, the atmospheric turbidity should be much lower in July than in the spring. We were also prepared to check this by both ground-based and aircraft measurements.

The field experiment in Barrow took place from 25 June through 22 July 1977, and was performed by Mr. Borys and Dr. Schütz of URI. The first three weeks were devoted to installing the aircraft air-sampling and radiation monitoring system, as well as collecting ground-level radiation data. The last week was a series of 5 flights, during which aerosols and gases were collected, radiation parameters were measured, and final checks of the system were made. During this last week ground-level radiation was also measured.

On each flight four sampling probes were used, for condensation-nucleus counts, Nuclepore and Millipore filters, and high-volume cellulose filters. The hemispheric flux monitor was run continuously whenever the sky was clear, and hand-held sun-photometer measurements were made at several altitudes depending on conditions.

July 1977 was abnormally clear for high summer in the Arctic. The majority of the time the skies were clear enough for radiation measurements to be taken, as opposed to the normal July which is dominated by low stratus clouds in Barrow. To the eye the Barrow atmosphere appeared deep blue and unusually clean, i.e., the turbidity values seemed very low, in particular much lower than the two previous springs. The radiation measurements, which are the only data we have available so far from this experiment, bear out these visual impressions. Indeed, the turbidities at Barrow in July 1977 were the lowest ever recorded there. Typical optical depths were 0.05 at the violet end of the spectrum and 0.01 - 0.02 at the red end. To have such low values at a sea-level site is rare indeed (only 5% of the violet radiation and only 1-2% of the red radiation interacted with the aerosol on its way through the atmosphere). The slope of the optical depth vs. wavelength profile was unusually great, that is, the Barrow aerosol of July 1977 seemed particularly depleted in giant particles ($r > 1$ μm). This also could be confirmed visually: when a thumb at arm's length was used to block out the disk of the sun, no aureole could be seen. This indicated the virtual absence of giant particles, which preferentially scatter sunlight in the forward direction.

Why should the turbidities at Barrow have been so low during July 1977? In general, we expected lower turbidities than in the spring, for several reasons. (1) McCall Glacier in northeastern Alaska shows summer turbidities several times lower than in the spring (see Figure 28 of Section ID). (2) Barrow itself has some evidence of a similar trend toward low turbidities in the summer (same figure), but the data are not nearly so conclusive. (3) We have hypothesized that the high spring turbidities in the Arctic are linked with pollution aerosol. If this is correct, the low pollution levels of summer ought to be accompanied by correspondingly low turbidities. Thus we

were not surprised to find low turbidities during July.

More specifically, the extremely low values of the turbidity during July appear to have been associated with a period of very low north-south exchange of aerosol in the Northern Hemisphere. July 1977 had a typical high-summer meteorological situation, when the polar front/jet stream was far to the north (close to 50°N over the US and Canada) and had very few waves. These "flat" contours reduced meridional exchange and therefore cut off mid-latitude pollution aerosol from the North. This was also the meteorological situation that created the intense heat wave of 14-23 July 1977 that affected almost all of the United States. The jet stream was located over southern Canada this whole time, with almost no waves in it. Hot, moist southern air spread over nearly all of the U.S. Because the jet was so far to the north, there was almost no pressure gradient across the United States, with the result that the overall circulation was very weak, and surface temperatures quickly built up to record levels. It is interesting that the same jet stream/polar front barrier that created heat, humidity, and high pollution levels in mid-latitudes also allowed the Arctic air to become unusually clean at the same time. Thus during July when the meteorological barrier was in place the north-south pollution contrasts increased. This interpretation of the turbidities of July 1977 confirms our picture of the jet stream/polar front as a highly effective barrier to the transport of pollution aerosol.

On a more local level, turbidity measurements at Barrow for 7-15 July for short wavelengths showed minimum values on the 12th and 13th, when Barrow was under the influence of a strong ridge of stagnant air. Before and after this period, when upper-level winds came from the south and north to northeast, respectively, the turbidities were higher. Interestingly, the optical depths at the red end of the spectrum, which reflect the abundances of larger

Particles than the violet end, showed no time trend.

Details of this experiment will be reported next year.

I. Elemental composition of desert dust as a function of particle size

The principal project of Dr. Lothar Schütz while he is spending 9 months as a visiting scientist with our group at URI is to use neutron activation to analyze several desert soils from the Sahara, Texas, and Arizona as a function of particle size. The purpose of this project is to compare the chemical composition of the bulk soil, which is often used as a reference material for calculation of aerosol-crust enrichment factors, with that of its aerosol-sized fraction. If systematic differences should be found, this would mean that the composition of the aerosol size range of the soil should be used as the reference material rather than the bulk soil. Of particular interest is the composition of the soil fraction with radius < 1 μm. These particles, when airborne, can travel thousands of kilometers, and represent the "background" crustal aerosol in the atmosphere. There already is evidence (Rahn, 1976) that the Si/Al ratio of remote aerosol is roughly a factor of two lower than either bulk soil or bulk rock, and we hope by this study to determine how many other elements are systematically fractionated.

The soil samples were size-fractionated at the Max-Planck-Institut für Chemie in Mainz, West Germany, Dr. Schütz's home institute. This work took more than 6 months, and represented a considerable cost-free contribution to our project. Methods used were dry sieving and sedimentation in water. In the ideal case a given soil sample could be fractionated into size ranges of roughly a factor of two, from a maximum radius of 400 μm to a miminum of less than 1 μm. Certain soils, however, had so little mass smaller than 8 or 16 μm radius that they could not be fractionated further.

Neutron activation analysis of these samples is under way. Detailed results will be reported next year. Already, though, a number of general first results are becoming apparent, which will now be briefly cited:

(1) By short (5-minute) irradiations alone some 16 elements are detectable (Si, Al, Mg, Ca, Sr, Ba, Ti, Mn, V, Dy, Na, Cl, Br, I, Cu, In). Not all of these are detectable in all samples, however. Nine elements per sample are always detected. In addition to the above, K, La, and Sm are usually seen. These are not counted here because they will be routinely determined later with longer irradiations.

(2) There is generally a broad zone of nearly constant elemental concentrations from about radius 1 μm to about 20-30 μm. Between 20-30 μm and 400 μm concentrations generally decrease by 1 to 2-1/2 orders of magnitude. All seven soil samples studied so far showed this effect in greater or lesser degree. Interestingly, the magnitude of the concentration decrease seems to be a property of the soil, that is, a given soil has a tendency for all elements to decrease greatly, or moderately, or only slightly.

(3) For a given soil not all elements decrease to the same degree between 20 and 400 μm. Of the elements determined so far, Ba tends to be the least concentrated in the small sizes and Na tends to be the most concentrated there. Many more data will be forthcoming on this point, however.

(4) Something in the soils must increase in concentration between 20 and 400 μm to compensate for all the other elements decreasing. We feel that this component must be the diluent SiO_2. This was hypothesized earlier (Rahn, 1976), and seems to be confirmed here by our observation that the Si/Al ratio decreases greatly (about 10x) with particle size. So far our Si data is not very good for the small particle sizes, where it is near the neutron activation detection limit. We will attempt to improve the Si data by irradiating these soils under Cd.

(5) In the one sample that has been irradiated so far for the fraction $r < 1$ μm, striking differences were seen relative to all larger sizes of this

same soil. In particular, Cl, Br, I, In, and Cu seemed to be enriched in this fraction. These same elements are also enriched in the small-particle size range of atmopsheric aerosols, and we wonder now how much of the aerosol enrichment may be due to enrichment on small soil particles.

J. Arctic aerosol planning conference of April 1977

Soon after we realized that the atmosphere of the high Arctic was routinely polluted, we began to formulate ideas on what the principal pathways of this aerosol to the Arctic from mid-latitudes might be. The three major pathways seemed to be (1) northeastward from the northeastern United States, veering northwestward around the southern tip of Greenland; (2) northeastward from the northeastern United States, over Iceland and Spitzbergen; and (3) northward or northwestward from Europe. We also realized that it might be possible to form a near-ideal large-scale network to monitor the flow of polluted air masses, using the conveniently located land masses of Greenland, Iceland, Spitzbergen, Jan Mayen Island, Bear Island, etc., which form a natural grid with spacing suitable for air sampling along these pathways.

In a discussion between Dr. B. Ottar, director of the Norwegian Institute for Air Research (NILU), and Dr. K. Rahn, of this project, held in October 1976, Dr. Rahn explained his ideas about long-range transport of aerosol to the Arctic. Dr. Ottar, who has been very interested in Arctic air chemistry for many years, noted that several of the sites on the proposed grid already had small air-sampling programs underway, operated by individual Scandinavian countries. Only some leadership was needed to coordinate these programs and to make this potential network an actuality. Drs. Rahn and Ottar decided to exert this leadership, and on 27 and 28 April 1977 a first meeting of countries bordering on the Arctic or concerned with Arctic air chemistry was held at the

Norwegian Institute for Air Research, Lillestrøm, Norway, supported by NILU and by a Conference Grant from ONR. The meeting was organized jointly between ONR and NILU, and was co-chaired by Drs. Ottar and Rahn with Mr. A. Semb as Rapporteur. There were 28 participants from 7 countries (Canada, Denmark, Finland, Norway, Sweden, the United Kingdom, and the United States). Iceland could not participate becasue of prior committments. The Soviet Union will be invited to participate in all future meetings.

The first day of the conference was devoted to presentations of ideas and results of Arctic research by the various participants. Opening keynote addresses were presented by Dr. Ottar ("Ideas for a measurement program in the Arctic region") and Dr. Rahn (" Air pollution in the Arctic - Some emerging ideas"). A series of 11 shorter presentations followed. Abstracts of all 13 presentations appear in the report of the conference, entitled "Sources and significance of natural and man-made aerosols in the Arctic," Norwegian Institute for Air Research, Lillestrøm, 21 June 1977.

The second day of the conference was devoted to a discussion of future research cooperation in the Arctic. The major conclusions of this discussion were: (1) Coordinated sampling and analysis of the Arctic aerosol is highly desirable because of a general concern for this environment. (2) A general, scientific study of atmospheric transport of aerosols to the Arctic is now warranted, in light of the recent evidence that large quantities of natural and pollution aerosol are carried into the Arctic from outside. (3) The possible climatic effects of these extra-Arctic aerosols need to be investigated for both direct (radiative) and indirect (alteration of cloud and precipitation patterns) effects. (4) Baseline studies of aerosol concentrations and composition in the late 1970's need to be performed immediately, because human activity in the Arctic is rapidly increasing.

Each of the organizations and/or countries attending the conference agreed to work separately on developing its own Arctic aerosol research program until

January 1978. At this time 3 or 4 representatives will meet at the Danish Air Quality Laboratory in Risø, to review the results, to assess the resources available, and hopefully to chart a course for future coordinated activities. A second, larger meeting is foreseen for April 1978. Prior to the January 1978 meeting, Dr. Rahn will be receiving and evaluating progress reports.

As is obvious from the above discussions, Dr. Rahn will play a very active role in the development of any Arctic air-sampling network. The existence of such a network should greatly facilitate our continuing ONR Arctic studies, and we trust that ONR will encourage us in this role. As will be discussed in detail in Section IIAld, we are already reaping the benefits of this conference, with aerosol samples becoming available to URI from Greenland and the Norwegian Arctic during late 1977.

II. Renewal proposal: 1 October 1977 - 30 September 1978

A. Introduction and rationale

Our work for the past 18 months has been unusually productive (see Section IA for highlights of the results). A number of clear-cut major ideas have emerged - the importance of Asian desert dust to the world atmosphere and to the Arctic atmosphere in particular; the presence of pollution aerosol in the Arctic; and the meteorological controls on transport of pollution aerosol to the Arctic from mid-latitude sources. In addition, there have emerged a number of less clear-cut but equally important ideas, such as whether there are indeed two major origins for Arctic haze, and the possible climatic effect of Arctic haze.

All of these ideas have one thing in common - they are still based on very little, usually incomplete data. For example, our ideas about meterorological controls on transport of pollution to the Arctic come from 7 months of data at Barrow, 5 months of data from Spitzbergen, and slightly over one year of data from northern Norway.

We feel very strongly that before these important ideas can be completely understood or accepted, they simply must be supported by a broader data base. For this reason a major part of our planned work for FY 78 is to continue the same type of experiment as we did in FY 77. In particular (1) we plan a spring 1978 intensive aircraft and ground campaign at Barrow, to confirm the presence of Asian dust there (with an expanded aerosol sampling system which will be supplemented by trace-gas measurements by Dr. John Kelley of NARL); (2) we will continue ground sampling of aerosol at Fairbanks and Barrow, to verify the routine presence of polluted air during the winter at Barrow, and also to verify the surprising result that the aerosol is significantly less polluted at the same time in Fairbanks;

(3) we will also continue the radiation measurements on the ground and aloft, which have a longer history at Barrow than do the chemical measurements.

Mere continuation is not enough, however. During FY 78 we will actively seek to refine the major hypotheses which we have been able to formulate only roughly during FY 77. Our work will include the following: (1) Concerning Asian dust, we will attempt to get better estimates from our chemical, physical, and optical data on the total concentration of Asian dust over Alaska. We will also cooperate with Dr. Ross Heath of URI, who is studying quartz in Pacific pelagic sediments, to derive an estimate of the total annual dust output from the Asian deserts. When this figure is compared with the value of 260×10^6 tons per year for the Sahara (see Section IF), we will have a much better idea of the relative importance of the Asian deserts in global atmospheric chemistry. We will try to arrange to set up an aerosol monitor in Kunsan, Korea. One of the results of this program should be to derive another estimate of the annual amount of Asian dust passing out over the Pacific, which can then be compared with the value based on deep-sea sediments, which is of course an integrated value over a very long time span rather than a current value.

(2) Furthermore, we will try to formulate more precisely our hypothesis of meteorological constraints on long-range transport of pollution aerosol to the Arctic. The proposed barrier effect of the jet stream/polar front system will be better calculaced, and correlations between pulses of air pollution at Barrow and specific meteorological flow patterns will be sought. Numerical calculations of winter-summer ratios of pollution concentrations in the Arctic, which Dr. Rahn will present in their present elementary form at the Ninth International Conference on Atmospheric Aerosol, Condensation and Ice Nuclei (21-27 September 1977, Galway, Ireland), will be

refined. Many more trajectories will be calculated between suspected source areas and the Arctic, to check our preliminary ideas about these principal atmospheric pathways between mid-latitudes and the north. (3) The most important hypothesis that the more diffuse Arctic haze (what we call Type B) is largely derived from air pollution, either directly or indirectly, will be further examined in light of all available information.

And lastly, during the next year we will take full advantage of a number of opportunities already afforded by the planned Arctic Network to obtain preliminary aerosol data from the eastern Arctic (between eastern Canada and Norway). Aerosol samples from Spitzbergen, Bear Island, and northern Norway will be supplied by Dr. B. Ottar of the Norwegian Institute for Air research, and samples from northern and southern Greenland will be supplied by Dr. Hans Buch of the Danish Meteorological Institute, Copenhagen. Dr. G.E. Shaw will use Dr. Ottar's aircraft and his own radiation instruments to obtain vertical aerosol profiles in the Norwegian Arctic. This radiation data will be the first from the eastern Arctic; the chemical aerosol data from the various filters will be confirmatory for Spitzbergen and northern Norway, but the first for Bear Island and Greenland. All these cooperative projects are the first fruits of the open communications associated with the proposed Arctic Network. The filter samples will cost this project nothing; the aerosol vertical profiles will be sponsored jointly by ONR and our new NSF proposal (see Section II B3).

In summary, we feel that this coming year of active but not totally new work is the most efficient way to refine our rapidly emerging ideas about the Arctic aerosol and its sources.

B. General plan of the work

1. Ground stations

a. Barrow - The chemical data drom the surface filters at Barrow have been of extremely high quality and were absolutely essential to the formulation of our hypothesis of meteorological control of long-range transport of pollution aerosol to the Arctic. One year ago we didn't know whether this particular experiment would have much value at all - in fact, it was foreseen as a way of measuring seasonal variations of Asian dust in the absence of aircraft flights all winter. As it turned out, the significance of the data was much broader than we had anticipated. and in a rather different direction. (This has been characteristic of our Arctic research so far - with surprising results of far-reaching importance.)

The ground samples were all taken for us by staff personnel of NOAA's GMCC (Geophysical Monitoring for Climatic Change) clean-air observatory in Barrow, for which we are greatly indebted. Dr. Kirby Hanson, director of the GMCC program, has approved a second year of sampling for us at Barrow. We will gladly continue, but with sampling times reduced to 4 days instead of one week, the period used during most of the 1976-77 season. This change was made because the 1976-77 data showed a series of pulses in concentration at Barrow, which seemed to be significantly better defined during a few periods when the sampling duration was shorter than a week.

Our specially designed pump-filter-shelter system worked very well in Barrow. The heat lamps kept frost off the filters and kept the anemometer system ice-free as well. The electronic control system and a small Gast vacuum pump, however, had to be replaced.

We have also decided to try to get a series of radiation measurements of atmospheric turbidity at Barrow for as much of the year as the

sun's disk can be seen. Dr. John Kelley of NARL has offered to have one of his technicians use Dr. Shaw's twelve-wavelength sun photometer on all clear days. Of particular interest at first will be the October - November period when pollution aerosol increases so rapidly. If the turbidity should increase simultaneously, this would be a major piece of evidence linking the two.

b. Fairbanks

The Fairbanks site was a major disappointment during FY 77. Located on Ester Dome, it had enough locally generated soil dust during dry snow-free periods to obscure any occurrence of the Asian dust that we were seeking. The highly unusual winter of 1976-77 created less-than-normal snowfall in Fairbanks and much warmer-than-normal temperatures. Together, these effects produced less than one month of snow cover on Ester Dome, as opposed to the normal several months. During most of the winter, therefore, we were not able to measure Asian dust at Fairbanks.

In spite of this setback, the Fairbanks site did provide some key data for the nature of its aerosol, and we will continue the sampling next year. Concentrations of noncrustal elements in the aerosol such as Cl, Br, I, As, Sb, Se, Cu, Zn, etc., were not disturbed by the local soil dust. About the first of November 1976, when vanadium and several other pollution elements sharply increased at Barrow, they did not increase at Fairbanks. This fact was instrumental in leading us to the conclusion that the increase at Barrow was primarily an Arctic phenomenon (Fairbanks is sub-Arctic). The pollution aerosol with these high concentrations came to Barrow from the north, and could not reach Fairbanks because of the shielding effect of the Brooks Range.

In fact, it was this striking contrast in aerosol character between

Fairbanks and Barrow that led us to propose a pollution origin for Type B Arctic haze. As noted in Section ID of this report, atmospheric turbidities at Barrow are routinely higher than at Fairbanks, when one would expect the opposite. When we found that pollution aerosol was also more concentrated in Barrow (and proposed a meteorological hypothesis which has since not yet been contradicted by any new evidence), we remembered that turbidity was also higher at Barrow, and started to think whether the two effects might be related.

We would like to confirm the Barrow-Fairbanks contrast for a second year by continuing sampling at Fairbanks. We will, however, attempt to relocate the Fairbanks site to a location free of local soil or pollution influences. This will not be easy, because we took quite some care in selecting Ester Dome in the first place. When Dr. Shaw returns to Fairbanks from Switzerland in mid-September 1977 this will be one of his first tasks.

c. Korea

The annual amount of Asian dust entering the Arctic atmosphere is a very important quantity, and one which we would like to be able to estimate. To do this, we need to know the seasonal frequency of dust events such as we observed in Barrow during April and May 1976. This information is not presently known. The seasonal pattern of Asian dust entering the Alaskan atmosphere is the product of the seasonal source function of the Asian deserts and the seasonal transport function to Alaska. The transport function can be easily seen from weather maps to be at a maximum during the winter half-year in general and during February in particular. The source function of the deserts remains elusive, however. The writers know of no published scientific studies of the seasonal frequency of Asian dust passing out over the Pacific. We have attempted to deduce this function from popular accounts of westerners who have lived in China, narratives of

expeditions across the Gobi Desert, etc., with only limited success and conflicting results.

A much more direct method to get this information is to monitor the source itself from somewhere just off the continent. We now have a chance to do this through ONR: Dr. Johnson of the Arctic Program, our project manager, has requested permission for us to place an aerosol sampler (the same type as used at the ground stations in Alaska) at Kunsan Air Force Base in Korea. This site is right along the path of the major dust storm of April 1976 which ultimately reached Barrow and was observed by us there. Although this is the only surface dust storm which we have studied so far, we feel that it may well have taken a typical path. If we are granted permission to use Kunsan Air Force Base, the results of a year's sampling there should be most informative.

d. Other sites

As mentioned above, we will be able to obtain valuable aerosol samples from a number of other sites during FY 78, mostly in the eastern Arctic in connection with the proposed Arctic Network. Dr. B. Ottar of the Norwegian Institute for Air Research has just set up three sampling sites in Spitzbergen, Bear Island, and northern Norway (Finnmark), to take continuous weekly samples indefinitely. He has offered us these samples for neutron activation of trace elements, by the same program that we have been following for the Barrow and Fairbanks samples. Inasmuch as these three sites represent Arctic, intermediate, and sub-Arctic climates, respectively, trace-element data for a year or more will greatly strengthen our data base for understanding the effect of the polar front on abundance of pollution aerosol in remote northern regions.

In addition, Dr. Hans Buch of the Danish Meteorological Institute has

offered us a shorter series of samples from two sites in northern and southern Greenland during summer and possibly also fall and winter 1977. The surface atmosphere around Greenland is of particular interest to us because of the discrepancy between our ideas of ubiquitous vanadium pollution in the Arctic atmosphere during winter and the apparent lack of pollution vanadium in recent Greenland ice from 2500 m elevation. (This discrepancy is discussed in detail, together with references, in Section IE of this report). One possible resolution of this conflict is that the pollution vanadium is confined to a relatively shallow surface layer of the atmosphere so that its abundance at 2500 m could easily be masked by layers of crustal dust, possibly from Asia. Surface aerosol samples from Greenland can begin to check this hypothesis, therefore we are interested in all the surface samples we can possibly get from the general vicinity of Greenland.

Lastly, it now seems possible to get an inexpensive, perhaps even cost-free, set of aerosol samples from one or two sites in the northeastern United States source region. One series could come from midtown Manhattan in New York City, on the roof of the 14-story New York University Medical Center student dormitory and classroom building. Dr. Theo J. Kneip of NYU's Environmental Medical Center maintains an active air-sampling station there, and is willing to help us change filters. A weekly or twice-weekly program would provide continuity with a set of weekly samples collected from July 1976 - June 1977 for Dr. Rahn's NSF halogen grant, the data from which can be used very nicely in conjunction with this Arctic program. In addition, a semi-weekly sampling program could easily be established at URI's Narragansett Bay Campus in Narragansett (a small town whose aerosol should be representative of the Northeast), where pumps and a shelter already exist.

This source-region data will be relevant to this project in a number of ways: (1) The seasonal pattern of pollutant concentration in the Arctic depends partly on the seasonal pattern of pollutant concentration at the source. We have a rough idea of seasonal trends of vanadium at New York City and Narragansett, but at both locations only a few other elements were determined, and at Narragansett in particular the sampling period was less than a year and was not continuous. A continuous series of samples from both places would therefore be highly desirable. (2) It may be possible to correlate weekly or semi-weekly pollutant concentrations in New York City with large-scale meteorological phenomena such as the locations of upper-atmosphere Rossby waves. Because the position of these same Rossby waves is what determines much of the long-range flow of pollutants to the North, correlation of wave patterns with concentrations at the sources could be a valuable first step in predicting the concentrations and regions of impact of this pollution far downwind.

2. Barrow 4-week campaign of spring 1978

We are planning a 4-week intensive experiment at Barrow for sometime in spring 1978, preferably March. It will be essentially the same as the experiment originally set for spring 1977 (which suffered a series of postponements until it became a July 1977 experiment), but with a few new wrinkles. The basic experiment will be a three-week series of flights in the C117D remote-sensing aircraft (150 flight hours anticipated), during which aerosol and gas samples will be taken and radiation measurements will be made. Aerosol samples will consist of high-volume samples on both cellulose and glass-fiber filters, for determination of chemical constituents and total mass. If possible, high-volume cascade impactor samples will also be taken, but we have been experiencing some severe problems with our Sierra

impactors under low-humidity conditions which for the moment preclude this kind of sample. In addition, our standard low-volume filters will be taken for ice-nucleus determinations (by Mr. Borys at Colorado State University) and scanning electron microscopy.

An addition to our program will be determination of trace gases by Dr. John Kelley of NARL. Dr. Kelley is an enthusiastic supporter of our program and will make gas measurements on all our flights. At the moment he measures CO_2, CO, H_2, light hydrocarbons, and chlorofluorocarbons (Freons), and has requested an instrument for O_3 from ONR. The addition of these gases to our flights should greatly add to the value of our overall program.

It is possible that Mr. Borys will be able to borrow continuous-reading ice-nucleus and/or cloud condensation-nucleus counters from Colorado State to use for this experiment. Details of these instruments are given in Section IIB5.

As in the July experiment, the radiation measurements will be conducted with a hand-held sun photometer and a hemispheric diffuse flux sensor mounted on the rear top of the fuselage.

The flights will have several purposes. They will be designed to check a number of aspects of both desert dust and pollution aerosol in the Arctic atmosphere. A long flight will be made following a haze band southward out over the Pacific Ocean if possible, to verify that the bands cannot originate within the State of Alaska itself. Vertical profiles of aerosol composition, as well as of turbidity, will be made in the vicinity Of Barrow. Prudhoe Bay will again be checked as a source for vanadium. Aerosol composition far north of Barrow will be studied under northerly winds, when the aerosol has a good chance of originating in Europe or North America.

All flights will be coordinated with ground sampling at the GMCC site in Barrow. During this experiment the standard 4-day ground samples will be replaced by daily ground samples, which can be related much better to the aircraft samples.

A second purpose of the spring 1978 experiment will be to install and test several minor modifications in the aircraft sampling system of the C117D. This sampling system is still very new - it was only installed in July 1977 and has been used on only five sampling flights. As a result of these, however, Mr. Borys and Dr. Schütz have recommended that several samll changes be made in both the probe assembly and the cabin equipment. This should take only a few days at Barrow.

The third and final aspect of the spring experiment will be a series of the following special experiments, some of which have nothing to do with the aircraft: (1) Aerosol samples will be taken just above the surface inversion layer. In principle one does not need an aircraft to do this, because the height of the inversion is about 100m in winter, but because our equipment is still rather bulky we will use the C117D. The object of this experiment is to see how representative surface air is of air just above the inversion. There is some concern that it may be less than perfectly representative. In Antarctica, for example, concentrations of aerosols seem to be measurably lower within the inversion layer than above it. Also, if the surface inversion layer turns out to have a different composition than the air just above, local sources may be important.

The second and third special experiments will be in conjunction with Dr. Kelley of NARL. He is presently studying the exhumation of CO_2, methane and other light hydrocarbon gases from the tundra as a function of season, and feels that the Barrow area may be a considerable natural source of these gases. Vanadium, which has been a very important element for us in deducing

the transport of pollution aerosol to the Arctic, is of course ultimately derived from oil deposits similar to those found in the Arctic. We think that the major source of vanadium in Arctic aerosol is combustion of residual oil in mid-latitudes, and have not reckoned with Arctic sources so far. Could any gaseous vanadium be leaking from the ground along with the CO_2, etc, that Dr. Kelley studies? If so, we would have to revise our ideas on vanadium in the Arctic. We doubt it, because vanadium's compounds are not volatile, but there is a way to check this possibility. Dr. Kelley has a chamber dug into the soil near NARL, where he performs some of his gas-leakage measurements. The chamber is sufficiently large (several hundred cubic feet) so that we could take an aerosol sample there and analyze it for vanadium. This we plan to do when we are in Barrow next spring.

The other cooperative experiment with Dr. Kelley will be analysis of melt water from cores of pack ice. Pack ice is known to contain measurable amounts of crustal dust, presumably some or most of which is related to Asian deserts. We would like to obtain some cores from Dr. Kelley, melt off the outside ice to remove possible contamination from drilling, subdivide, melt and filter them, and return them to URI for analysis. We will be looking to see whether individual dust episodes can be detected as layers in the ice, whether pollution products can also be seen in the ice, and to what extent a removal rate for Asian dust can be calculated. This last information will be used as a first estimate of the fate of Asian dust in the Arctic atmosphere, a topic which will eventually be of great interest to us.

3. Aerosol vertical profiles from the Norwegian Arctic

One of the most important properties of Arctic aerosols is their vertical distribution. Climatic effects of aerosols, for example, depend not only on their concentration and particle-size distribution, but also

on their vertical distribution. The vertical profile of aerosol composition is also very important and virtually uninvestigated in the Arctic. The disagreement between our high surface vanadium concentrations in the Arctic and the low pollution concentrations in Greenland ice suggests that the aerosol near Greenland may change radically in composition with height within the first 2500 m.

The only vertical distributions of aerosol for the Arctic are for the vicinity of Barrow and were measured by Dr. Shaw on this and previous projects. We would like to obtain similar information for the eastern Arctic as part of this year's work. Dr. B. Ottar of the Norwegian Institute for Air Research, Lillestrøm, has offered us the use of his Piper Aztec twin-engine aircraft for $200 per hour, all expenses included. This plane can cruise for five hours, which gives it an effective range of northern Norway to Bear Island and back, or northern Norway to Spitzbergen one-way. We envision an introductory experiment only, of approximately two weeks' duration, with 3-4 good flights of about 5 hours each. The best time for this would be in late spring, when the chances of having clear weather in the Norwegian Arctic are the greatest. Dr. Shaw himself will travel to Norway and make the measurements with his hand-held, twelve-wavelength sun photometer. As noted in the budget, we have allowed $5000 for this experiment, to be divided equally between this project and our NSF proposal. If this experiment is successful, we will probably continue it in the future.

4. Meteorological analysis

Our next year's work would not be complete without continuing attention being paid to improving our understanding of the meteorological aspects of transport of Asian desert dust and European and American pollution aerosol to the Arctic.

Concerning transport of desert dust, we will obtain radiosonde data for Asia, Japan, Korea, the Pacific, and Alaska for April and May 1976, our first spring experiment. These data will be plotted and inspected for clues about the transport height of dust bands. Dr. Schütz, who will be working with us at URI until mid-December 1977, is particularly interested in looking into this topic. Similar analysis for Sahara dust transport was an important part of his Ph.D. thesis in Mainz, West Germany (see Section IF). After 1 October 1977 he will be working for this project at no cost, his salary coming from the Max-Planck-Institut für Chemie, Mainz.

Concerning transport of pollution aerosol, we will construct a large number of forward and backward trajectories between North America and European source areas and the Arctic receptor area. We hope that this will ultimately give us a better feeling for the prinicpal pathways of atmospheric transport to the Arctic, as well as the general sources of the aerosols of northern Norway, Spitzbergen, Iceland, and Greenland. Later when the Arctic Network begins to operate, we hope to be able to track polluted air masses directly.

If time permits we will investigate the feasibility of using other indicators of air-mass transport, such as temperature, vertical structure, etc. There is already some indication that a "blob" of cold air can be followed by its temperature alone as it passes over the pole from northern Europe to Barrow. These possibilities are definitely worth following up.

5. Ice-nucleus analysis

Mr. Randolph D. Borys, who has been associated with this program since its inception as a Research Assistant at URI, has recently enrolled as a Ph.D. student in the Department of Atmospheric Science of Colorado State University. Because of Mr. Borys' deep interest in this work, URI and CSU have worked out an arrangement whereby he will continue to work on

this project as his thesis. As noted in the budget, we will pay one-half his graduate stipend ($3000 per year) from this project for the first year, perhaps more later. In return, he will have as his thesis topic the physical effects of natural and pollution aerosols on the Arctic atmosphere, with particular reference to aerosols originating outside the Arctic which have been transported in. His main area of concern will be ice nuclei and whether those from outside the Arctic exert a measurable effect on the precipitation processes or cloud cover of the Arctic. Should the answer to either of these questions be positive, these effects would have to be considered in future climatic studies of the Arctic. We now know that large amounts of both natural and pollution aerosol are being carried into the Arctic atmosphere each year; because both these types are known to be good ice-nucleating agents, we have good reason to expect that Mr. Borys' measurements will be significant.

Mr. Borys may also attempt to measure activity spectra of cloud-condensation nuclei in the Arctic. This type of measurement is still incompletely understood and subject to considerable controversy, however, and the fate of this aspect of Mr. Borys' work is still very uncertain.

Two basic types of samples will be taken - Millipore filters which will be analyzed later at CSU, and samples analyzed in situ during the flights. Millipore samples from the spring 1976 and July 1977 experiments already exist and will be analyzed soon, hopefully before the next spring experiment. Additional Millipore samples will also be taken during the March 1978 experiment and possibly also as part of the Arctic Network. In situ analysis is hoped to be a part of the March 1978 experiment, but the availability of the instruments from CSU is not yet certain.

The basic instrument available for analysis of the Millipore filters is a Meeda static thermal-diffusion chamber, which activates ice nuclei. Mr.

Borys will begin his work by performing a series of checks of its reproducibility on local aerosol samples. If this succeeds, the existing Millipore filters will be analyzed. If this check fails, it may be possible to use the instrument of G. Langer of NCAR, the results from which are generally accepted.

The other ice-nucleus counter which we hope to use in the program is a continuous recording instrument, the Mee Continuous Ice Nucleus Monitor Model 140. It works, it has been flown by CSU, requires only 24V DC aircraft power and some dry ice for a cold sink. It needs recalibration, however, because the electronics have been modified. It will be used for about 5 weeks on the NCAR Queen Aire in December 1977 or January 1978 and should be available for our use in March 1978. It operates by humidifying the air, cooling it to form a cloud, then supercooling the cloud until any ice nuclei are activated and form crystals. These crystals are then counted by laser light scattered by the crystals as they fall past a detector. This is a very realistic way of counting ice nuclei.

Also available is a Mee cloud condensation nucleus counter, which is also set up for aircraft use. It operates much like a normal condensation nucleus counter, but at much lower supersaturations (less than 1%). We should be able to get activity spectra with this instrument, which would be important for getting an idea of potential cloud-droplet size spectra which could be incorporated into future radiation calculations for the Arctic.

6. Chemical analysis of desert soils as a function of particle size

Dr. Schütz is well under way in determining the chemical composition of a suite of desert soils as a function of particle size from radius 400 μm down to radius < 1 μm. During the last three months of his visit at URI (October - December 1977) he will conclude this work, which is

expected to result in a wealth of information about the detailed composition of the crustal component of the atmospheric aerosol. For example, he has 10 soil samples and approximately 80 size-segregated fractions. Analysis of each fraction for 30 elements will yield a total of 2400 data points.

The FY 78 portion of Dr. Schütz' time at URI will be financed by his home institute in Mainz, West Germany.

7. Specific timetable

October - December 1977

Analyze Fairbanks, Barrow, Spitzbergen, Bear Island, Northern Norway, and Greenland ground aerosol samples. Analyze July 1977 Barrow aircraft samples. Complete analyses of desert soil samples. Prepare for spring 1978 Barrow experiment. Meteorological analyses.

January - March 1978

Attend Arctic Network technical meeting (Rahn). Continue analyses of ground aerosol samples. Final preparations for spring experiment. Perform spring experiment.

April - June 1978

Attend second large Arctic Network meeting (Rahn, Shaw, depending on funding). Continue analysis of ground samples. Begin analysis of Barrow spring experiment. Perform Norwegian Arctic radiation experiment.

July - September 1977

Analyze ground samples. Finish analysis of Barrow spring experiment. Analyze data from Norwegian Arctic radiation experiment.

C. Facilities available

University of Rhode Island

Two well-equipped chemistry laboratories, one in the Horn Building at the Graduate School of Oceanography and the other in the Rhode Island Nuclear Science Center (RINSC) adjacent to the School of Oceanography, will be used in this work. A new $30,000 Class 100 clean laboratory has just been constructed for our group in the RINSC and should be available by September 1977. Also available for this project are three laminar-flow clean benches. A machine shop is located on the main campus.

All neutron activation analysis will be performed at the RINSC. The RINSC swimming pool reactor operates at two megawatts and delivers a thermal neutron flux of approximately 4×10^{12} n $cm^{-2}sec^{-1}$ at the irradiation sites. Counting equipment available for this work includes several large Ge(Li) solid-state detectors and associated electronics, which may be used with either a Nuclear Data Model 2200 4096-channel analyzer or a Digital Equipment Corporation PHA-11 pluse-height analysis system. Data and spectrum reduction on these systems can either be done via magnetic tape output with subsequent use of the IBM 370-65 computer on the main campus or directly on the PDP-11/40 computer which is part of the PHA-11 system.

Also located at the RINSC is a complete atomic absorption analysis laboratory, including three Perkin-Elmer instruments (Models 360, 503 and 603), HGA-2000 and HGA-2100 heated graphite atomizer attachments and other accessories.

The Graduate School of Oceanography has a Stereoscan S4 scanning electron microscope (Cambridge Scientific Instruments, Ltd.) which is available for general use. It features two CRT displays, a magnification

range of 10-200,000 X and a resolution in the secondary election imaging mode of 150A at 30 kV. In addition, the GSO has recently acquired a JEOL 50-JXA electron microprobe with 3-wavelength spectrometers and the KRISEL automation package, including a PDP-11/05 16K computer. This unit has 70-A resolution and a 1-μm beam.

University of Alaska

The Geophysical Institute of the University of Alaska is housed in the eight-story C.T. Elvey Building on the West Ridge of the College campus. Available facilities here include machine shops, welding, carpentry, painting, staging and assembly, electronics shop, instrument repair, electron microscope laboratory, optics dark tunnel, photographic services, drafting, reports production, programming and computing, data processing, optics laboratory, meteorology laboratory and optics test laboratory.

The Geophysical Institute library offers a comprehensive coverage of solar-terrestrial physics, meteorology and climatology, glaciology, oceanography, the solid-earth sciences, and environmental studies. The archives, adjacent to the library, include a world-wide collection of auroral and magnetic records, together with a more selective sampling of other records in the various geophysical disciplines.

Colorado State University

The Department of Atmospheric Science of Colorado State University is large and well-equipped for a variety of atmospheric investigations. The facilities which we plan to use for this project are ice-nucleus and condensation-nucleus counters, including: a Meeda static thermal-diffusion chamber, a Mee Continuous Ice Nucleus Monitor Model 140, and a Mee cloud condensation-nucleus counter. The first of these remains in the laboratory and is used for analysis of Millipore filters taken in the field, and the last two instruments are used in aircraft for direct readings during the flights. In addition to the above, there are two other ice-nucleus counters which are used for calibration purposes.

D. CURRENT SUPPORT AND PENDING APPLICATIONS

Dr. K.A. Rahn

Title	Agency	Duration	Amount	P.I. Time
Arctic Haze: Natural or Pollution? (This proposal) N00014-76-C-0435	ONR	1 Oct 1976-30 Sept 1977 (renewal expected)	$61,762	6 months
Atmospheric chemistry of the Halogens: Natural and Anthropogenic ATM 75-23725	NSF	1 Jan 1976-31 Dec 1977	$90,364	6 months
Pending Application				
Climatically Important Properties of Arctic Haze	NSF	1 Jan 1978-31 Dec 1978	$41,196	6 months

Dr. G. E. Shaw

Arctic Haze: Natural or Pollution ? (This proposal) N00014-76-C-0435	ONR	1 Oct 1976-30 Sept 1977 (renewal expected)	$7,549	1 month
Studies of the Atmospheric Radiation Field in Antarctica DPP 76-20629	NSF	15 Nov 1976-30 Apr 1978	$30,500	4 months
Studies of Scattered Sunlight and the Twilight Sky ATM 77-04432	NSF	15 July 1977-31 Dec 1978	$55,500	5 months
Pending Applications				
Climatically Important Properties of Arctic Haze	NSF	1 Jan 1978-31 Dec 1978	$58,755	6 months
Radiative Transfer Calculations for the Jovian Atmosphere	NASA	1 year	$36,450	

Pending Application (continued)

Studies of Atmospheric Particulates and Atmospheric NO_2	NSF	1 year	$46,828

E. Budget - Fiscal Year 1978

	URI	UA
(1) Salaries		
Co-principal Investigator Dr. Kenneth Rahn Research Assoc. - 6 months	10,500	
Research Assistant, Borys replacement, 12 months	16,000	
Secretary, 2 months	1,400	
Co-principal Investigator Dr. Glenn E. Shaw Assoc. Professor, 2 months		6,520
10% FY 78 increment		650
Reserve for annual leave 12%		780
Holiday, sick leave 9.5%		620
TOTAL SALARIES	27,900	8,570
(2) Fringe Benefits 12%	-	1,030
(3) Permanent equipment		
1 Rotron high-volume pump	400	
1 Temperature-dewpoint sensor	1,000	
1 1000-watt inverter	1,500	
TOTAL PERMANENT EQUIPMENT	2,900	-
(4) Expendable equipment and supplies		
Miscellaneous laboratory chemicals, glassware, etc.	1,000	
Miscellaneous aircraft-sampling supplies	1,000	
TOTAL EXPENDABLE EQUIPMENT AND SUPPLIES	2,000	-
(5) Travel Domestic:		
RI - Barrow (3)	3,000	
Attend scientific conference (2)	1,000	

Domestic Travel (continued)

	URI		UA
Trips to Ester Dome to change samples			125
Fairbanks - Barrow (3)			500
CSU - Barrow (1)	700		
TOTAL DOMESTIC TRAVEL	4,700		625
Foreign travel			
Attend two planning meetings for Arctic Network (first in Denmark, second as yet unspecified but in Scandanavia), Rahn	2,500		
Set up sampling station in Korea	2,000		
Field work in Norway, spring 1977			1,000
TOTAL FOREIGN TRAVEL	4,500		1,000
TOTAL TRAVEL	9,200		1,625
(6) Publications	500		250
(7) Other:			
R. Borys, Colorado State Univ. ice-nucleus determinations	3,000		
Computer	750		
Maintenance of sampling and analytical equipment	900		
Freight	1,500		
Telephone	900		
Scanning electron microscope	250		
Electron microprobe	250		
Aircraft fees, Norway			1,500
Meteorological maps on microfilm	1,000		
TOTAL OTHER	8,550		1,500
TOTAL DIRECT COSTS	51,050		12,975
(8) Indirect costs			
Overhead @ 55% of salaries	15,345		
Overhead @ 64.7 % of salaries			5,545
TOTAL COSTS	66,395		18,520
GRAND TOTAL		$84,915	
Less funds remaining from FY 77		-5,000	
AMOUNT REQUESTED		$79,915	

F. Comments on the budget

We are requesting a total of $79,915 from ONR for FY 78, which is the difference between our actual FY 78 operating budget of $84,915 and our estimated surplus of $5,000 as of the end of FY 77. Of the total budget, $58,390, or about 70%, is salaries, fringe benefits, and overhead. Specific comments on the various sections follow.

(1) Salaries: We are requesting 6 months' salary for Co-Principal Investigator Dr. Kenneth A. Rahn of URI. This half-time level of participation is the same as it has been for the last year of this project. At the moment Dr. Rahn's other commitment is half-time to an NSF grant on atmospheric halogens (see Section IID), which ends 31 December 1977. It has not yet been decided whether to apply for renewal of this grant, and if so, at what funding level. In the event that renewal is not requested, however, Dr. Rahn, together with Dr. Shaw, has applied to NSF Division of Polar Programs for a one-year grant to study the climatically important properties of Arctic haze, which would begin 1 January 1977 and support him half-time.

We are also requesting 12 months' salary for an unnamed assistant to Dr. Rahn, to replace Mr. Borys, who is now a doctoral student at Colorado State University. We are now in the process of finding this replacement.

Two months' salary is also requested for secretarial support, a level which seems reasonable.

University of Alaska

We are requesting two months' salary for Co-Principal Investigator Dr. Glenn E. Shaw. This is double his official participation for FY 77, and more accurately reflects his actual participation in the project.

(3) Permanent equipment

Our permanent equipment requests are very limited this year. We wish to purchase a single Rotron high-volume pump as a spare because we anticipate sending our present spare pump to Korea. For the C117D sampling aircraft at NARL we have to purchase a single 1000-watt inverter, to pay NARL back for one which they bought for us this summer. In addition, we wish to add a temperature-dewpoint sensor to aid the sampling personnel in understanding the kind of air they are in so that they can better choose the sampling altitude during flight.

(5) Travel

We have budgeted three round trips between Rhode Island and Barrow for this year. The basic March 1978 experiment will require two persons; the third trip is as an emergency trip, such as we needed this spring when the sampling program had to be postponed after Mr. Borys had reached Barrow.

In view of the large number of results and ideas that we have generated in this project so far, we feel that attendance at two domestic scientific conferences is reasonable.

Three Fairbanks - Barrow trips are requested. This number is probably a mimimum, because we want to collect radiation data throughout the year at Barrow.

One trip for Mr. Borys from Colorado to Barrow is foreseen, for the spring experiment.

For foreign travel, we are requesting funds for Dr. Rahn to attend and preside over two planning meetings for the Arctic Network. Both these meetings will be held in Scandinavia. The first will be held in January 1978 at the Danish Air Quality Laboratory in Risø; the location

of the second meeting is not known. In view of the great benefits that ONR will (and in fact have already started to) gain from our association with this network, we feel that this investment is more than worthwhile. We hope that Dr. Shaw will be able to conduct his Norwegian Arctic radiation measurements just before or after this conference, so that he can attend at no extra cost.

We are also requesting funds for Dr. Rahn to set up the aerosol-sampling station in Korea. It is expected that he will be able to meet with Dr. Kenji Isono, the Japanses scientist who has studied ice nuclei associated with Asian dust, on this same trip.

Lastly, we are requesting half of Dr. Shaw's travel costs to Norway for the radiation experiment in the Norwegian Arctic. As noted in the text, our NSF proposal is expected to provide the other half of these funds.

(7) Other

We are requesting $3,000 as a partial graduate stipend for Mr. Randolph D. Borys at Colorado State University. This will support his work on ice nuclei in the Arctic atmosphere, data for which should be of great value to this project. The work will also be his Ph.D. thesis there.

The $1,500 aircraft fees are one-half what it will cost for 3 to 4 5-hour flights of Dr. Ottar's Piper Aztec for Dr. Shaw's vertical radiation profile experiment of spring 1978. The other half of the funding will come from our proposed NSF grant.

G. Biographical Information

Co-Principal Investigator

NAME: Kenneth A. Rahn

EDUCATION: B.S. Massachusetts Institute of Technology, 1962 (Chemistry)

Ph.D. University of Michigan, 1971 (Meteorology)

PROFESSIONAL EXPERIENCE:

1976 - Present	Research Associate, Graduate School of Oceanography University of Rhode Island
1975 - 1976	Invited Visiting Scientist Max-Planck-Institut für Chemie, Mainz, West Germany
1973 - 1975	Research Associate, Graduate School of Oceanography University of Rhode Island
1971 - 1973	Research Associate, Institute for Nuclear Sciences University of Ghent, Belgium
1968 - 1971	Graduate Assistant, University of Michigan Ann Arbor, Michigan
1970 - summer	Lawrence Radiation Laboratory, Livermore, California
1963 - 1968	Science and Mathematics teacher Classical High School and Barrington College Providence, Rhode Island

PROFESSIONAL SOCIETY MEMBERSHIPS: American Chemical Society
American Association for the Advancement of Science
American Meteorological Society

RECENT PUBLICATIONS:

Dams, R., J.A. Robbins, K.A. Rahn, and J.W. Winchester, "Non destructive neutron activation analysis of air pollution particulates", Anal. Chem., 42, 861 (1970).

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RECENT PUBLICATIONS:

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